Implementation and evaluation of the GEOS-Chem chemistry module version 13.1.2 within the Community Earth System Model v2.1

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Short summary. We bring the state-of-the-science chemistry module GEOS-Chem into the Community Earth System Model (CESM). We show that some known differences between results from GEOS-Chem and CESM's CAM-chem chemistry module may be due to the configuration of model meteorology rather than inherent differences in the model chemistry. This is a significant step towards a truly modular ESM and allows two strong but currently separate research communities to benefit from each other's advances.

Abstract. We implement the GEOS-Chem chemistry module as a chemical mechanism in the Community Earth System Model version 2 (CESM). Our implementation allows the state-of-the-science GEOS-Chem chemistry module to be used with identical emissions, meteorology, and climate feedbacks as the CAM-chem chemistry module within CESM. We use coupling interfaces to allow GEOS-Chem to operate almost unchanged within CESM. Aerosols are converted at each time step between the GEOS-Chem bulk representation and the size-resolved representation of CESM's Modal Aerosol Model (MAM4). Land type information needed for dry deposition calculations in GEOS-Chem is communicated through a coupler, allowing online land-atmosphere interactions. Wet scavenging in GEOS-Chem is replaced with the Neu and Prather scheme, and a common emissions approach is developed for both CAM-chem and GEOS-Chem in CESM.

We compare how GEOS-Chem embedded in CESM (C-GC) compares to the existing CAM-chem chemistry option (C-CC) when used to simulate atmospheric chemistry in 2016, with identical meteorology and emissions. We compare atmospheric composition and deposition tendencies between the two simulations and evaluate the residual differences between C-GC compared to its use as a standalone chemistry transport model in the GEOS-Chem High Performance configuration (S-GC). We find that stratospheric ozone agrees well between the three models with differences of less than 10% in the core of the ozone layer, but that ozone in the troposphere is generally lower in C-GC than in either C-CC or S-GC. This is likely due to

greater tropospheric concentrations of bromine, although other factors such as water vapor may contribute to lesser or greater extents depending on the region. This difference in tropospheric ozone is not uniform, with tropospheric ozone in C-GC being 30% lower in the Southern Hemisphere when compared to S-GC but within 10% in the Northern Hemisphere. This suggests differences in the effects of anthropogenic emissions. Aerosol concentrations in C-GC agree with those in S-GC at low altitudes in the tropics but are over 100% greater in the upper troposphere due to differences in the representation of convective scavenging. We also find that water vapor concentrations vary substantially between the standalone and CESM-implemented version of GEOS-Chem, as the simulated hydrological cycle in CESM diverges from that represented in the source NASA MERRA-2 reanalysis meteorology which is used directly in the GEOS-Chem CTM.

Our implementation of GEOS-Chem as a chemistry option in CESM (including full chemistry-climate feedback) is publicly available and is being considered for inclusion in the CESM main code repository. This work is a significant step in the MUlti-Scale Infrastructure for Chemistry and Aerosols (MUSICA) project, enabling two communities of atmospheric researchers (CESM and GEOS-Chem) to share expertise through a common modeling framework and thereby accelerate progress in atmospheric science.

1. Introduction

Accurate representation and understanding of atmospheric chemistry in global Earth System Models (ESMs) has been recognized as an urgent priority in geoscientific model development. The National Research Council (NRC) report on a National Strategy for Advancing Climate Modeling (Bretherton et al., 2012) stresses the need for including comprehensive atmospheric chemistry in the next generation of ESMs. The NRC report on the Future of Atmospheric Chemistry (NRC, 2016) identifies the integration of atmospheric chemistry into weather and climate models as one of its five priority science areas. This work responds to those needs, presenting the implementation of the state-of-science model GEOS-Chem (Bey et al., 2001; Eastham et al., 2018) as an atmospheric chemistry module within the Community Earth System Model (CESM) (Hurrell et al., 2013; Tilmes et al., 2016; Lamarque et al., 2012; Emmons et al., 2020).

GEOS-Chem is a state-of-the-science global atmospheric chemistry model developed and used by over 150 research groups worldwide (http://geos-chem.org). It has wide appeal among atmospheric chemists because it is a comprehensive, state-of-science, open-access, well-documented modeling resource that is easy to use and modify but also has strong central management, version control, and user support. The model is managed at Harvard by a GEOS-Chem Support Team with oversight from an international GEOS-Chem Steering Committee. Documentation and communication with users is done through extensive web and wiki pages, email lists, newsletters, and benchmarking. Grass-roots model development is done by users, and inclusion into the standard model is prioritized by Working Groups reporting to the Steering Committee. The model can simulate tropospheric and stratospheric oxidant-aerosol chemistry, aerosol microphysics, and budgets of various gases .

65 Simulations can be conducted on a wide range of computing platforms with either shared-memory (OpenMP) or distributed
66 memory (MPI) parallelization – with this latter implementation referred to as GEOS-Chem High Performance, or GCHP
67 (Eastham et al., 2018).

69 For the general atmospheric chemistry problem involving K atmospheric species coupled by chemistry and/or aerosol
70 microphysics, GEOS-Chem solves the system of K coupled continuity equations

$$\frac{\partial n_i}{\partial t} = -\nabla \cdot (n_i \mathbf{U}) + P_i(\mathbf{n}) - L_i(\mathbf{n}) \tag{1}$$

74 where $\mathbf{n} = (n_1, \dots n_K)^T$ is the number density vector representing the concentrations of the K species, $\mathbf{U}$ is the 3-D wind vector,
75 and P_i and L_i are local production and loss terms for species i including emissions, deposition, chemistry, and aerosol physics.
76 The transport term $-\nabla \cdot (n_i \mathbf{U})$ includes advection by grid-resolved winds as well as parameterized subgrid turbulent motions
77 (boundary layer mixing, convection). The local term $P_i(\mathbf{n})$ $-L_i(\mathbf{n})$ couples the continuity equations across species through
78 chemical kinetics and aerosol physics.

80 Standard application of the GEOS-Chem model as originally described by Bey et al. (2001) is off-line, meaning that the model
81 does not simulate its own atmospheric dynamics. Instead, it uses analyzed winds and other meteorological variables produced
82 by Goddard Earth Observation System (GEOS) simulations of the NASA Global Modeling and Assimilation Office (GMAO)
83 with assimilated meteorological observations. The near-real time GEOS Forward Processing (GEOS-FP) output provides data
84 globally at a horizontal resolution of 0.25°×0.3125°, and the Modern-Era Retrospective analysis for Research and Applications,
85 Version 2 (MERRA-2) provides data at 0.5°×0.625°. GEOS-Chem simulations can be conducted at that native resolution or
86 at coarser resolution (by conservative re-gridding of meteorological fields). Long et al. (2015) developed an on-line capability
87 for GEOS-Chem to be used as a chemical module in ESMs, with initial application to the GEOS ESM. In that configuration,
88 GEOS-Chem only solves the local terms of the continuity equation

$$\frac{\partial n_i}{\partial t} = P_i(\mathbf{n}) - L_i(\mathbf{n}) \tag{2}$$

92 and delivers the updated concentrations to the ESM for computation of transport through its atmospheric dynamics. On-line
93 simulation avoids the need for a meteorological data archive and the associated model transport errors (Jöckel et al., 2001; Yu
94 et al., 2018). It also enables fast coupling between chemistry and dynamics.

96 Transformation of GEOS-Chem to a grid-independent structure was performed transparently, such that the standard GEOS-
97 Chem model uses the exact same code for on-line and off-line applications. This includes a mature implementation within the

GEOS ESM. It was applied recently to a year-long tropospheric chemistry simulation with ≈12 km (cubed-sphere c720) global resolution (Hu et al., 2018), and is now being used for global atmospheric composition forecasting (Keller et al., 2017, 2021). However, the only implementations of GEOS-Chem which are currently publicly available are either designed to run "offline", driven by archived meteorological data from the NASA Goddard Earth Observing System (GEOS) (Bey et al., 2001; Eastham et al., 2018), or operate at regional scale and do not extend to global simulation (Lin et al., 2020; Feng et al., 2021).

Integration of GEOS-Chem as a chemistry option within an open-access, global ESM responds to the aforementioned calls from the NRC. One of the most widely used open-access ESM is the Community Earth System Model (CESM) (Hurrell et al., 2013). CESM is fully coupled and state-of-science. It produces its own meteorology based on fixed sea surface temperatures or with a fully interactive ocean model. It can also be nudged to observed meteorology including from GEOS. The CESM configuration with chemistry covering the troposphere and stratosphere is referred to as CAM-chem (Community Atmosphere Model with chemistry) (Tilmes et al., 2016; Lamarque et al., 2012). CAM-chem is a state-of-science model of atmospheric chemistry; it has participated (along with CESM's WACCM model which extends to the lower thermosphere) in many international model intercomparison activities such as ACCMIP, CCMI, POLMIP, HTAP2, GeoMIP and CMIP6, and has a large international user community. CAM-chem also has a very different development heritage from GEOS-Chem, with each model providing better performance in comparison to observations in different areas (Park et al., 2021; Emmons et al., 2015; Nicely et al., 2017; Jonson et al., 2018). It is widely used for simulations of global tropospheric and stratospheric atmospheric composition, in part because it is able to run either with specified meteorological datasets or with fully-coupled physics (https://www2.acom.ucar.edu/gcm/cam-chem).

The fundamental differences in implementation of almost every atmospheric process between GEOS-Chem and CAM-chem mean that it is difficult to disentangle the root causes of these differences. Modular Earth system models can resolve this issue. Allowing individual scientific components to be swapped freely allows researchers to evaluate exactly what effect that component has in isolation, while also giving a single user base access to a larger portfolio of options. If two different models each implement five processes in different ways, a researcher must learn to use both in order to compare their results and cannot isolate the effect of any one process with confidence. If process options are implemented in the same framework, this problem is avoided. Such modularity is becoming increasingly possible with the availability of Earth system infrastructure such as the Earth System Modeling Framework (ESMF) and the National Unified Operational Prediction Capability (NUOPC), which describe common interfaces for Earth system modeling components (Hill et al., 2004; Sandgathe et al., 2011). The Multi-Scale Infrastructure for Chemistry and Aerosols (MUSICA) builds upon this trend with process-level modularization, with the goal of allowing researchers to select from a range of community-developed options when performing atmospheric simulations.

This work integrates the GEOS-Chem chemistry module into CESM as an alternative option to CAM-chem. Our implementation allows researchers to select either model to simulate gas-phase and aerosol chemistry throughout the troposphere and stratosphere, while other processes such as advection, broadband radiative transfer, convective transport, and emissions are handled nearly identically. We demonstrate this capability by comparing simulations of the year 2016 as generated by GEOS-Chem and CAM-chem operating within CESM, with the chemical module being the only difference. Estimates of atmospheric composition are compared between the two models and against a simulation in the standalone GCHP chemistry transport model (CTM). Finally, we evaluate the accuracy of the three approaches against observations of atmospheric composition and deposition.

Section 2 provides a technical description of the implementation of GEOS-Chem into CESM. Section 3 then describes the model setup. Sections 4 and 5 present a one-year simulation (not including spin up) performed in CESM with GEOS-Chem; CESM with CAM-chem; and the standalone GEOS-Chem CTM. This includes model intercomparison (Section 4), and evaluation against surface and satellite measurements (Section 5).

2. Coupling between GEOS-Chem and CESM

We first describe the interface used within CESM when using either the CAM-chem or GEOS-Chem options (Section 2.1). Unless otherwise stated, "GEOS-Chem" refers to the grid independent chemistry module which is common to all implementations, including standalone GEOS-Chem with OpenMP (Classic) or MPI (GCHP) parallelization, NASA GMAO's GEOS ESM, and WRF coupled with GEOS-Chem (WRF-GC). We then briefly summarize the chemistry and processes represented by the CAM-chem and GEOS-Chem options within CESM (Section 2.2). This is followed by a description of differences between the implementation of GEOS-Chem in CESM and its stand-alone code (Section 2.3), differences in the data flow through CESM when using GEOS-Chem as opposed to CAM-chem (Section 2.4), and finally the installation and compilation process (Section 2.5).

2.1. Interface

Our approach embeds a full copy of the GEOS-Chem chemistry module source code (version 13.1.2) within CESM (version 2.1.1). All modifications made to the GEOS-Chem source code have been propagated to the GEOS-Chem main code branch (https://github.com/geoschem/geos-chem) to ensure future compatibility between CESM and GEOS-Chem. Information is passed between the CESM Community Atmosphere Model (CAM) version 6 (CAM6) and the GEOS-Chem routines through an interface layer developed as part of this work. A schematic representation of the implementation is provided in Figure 1.

161

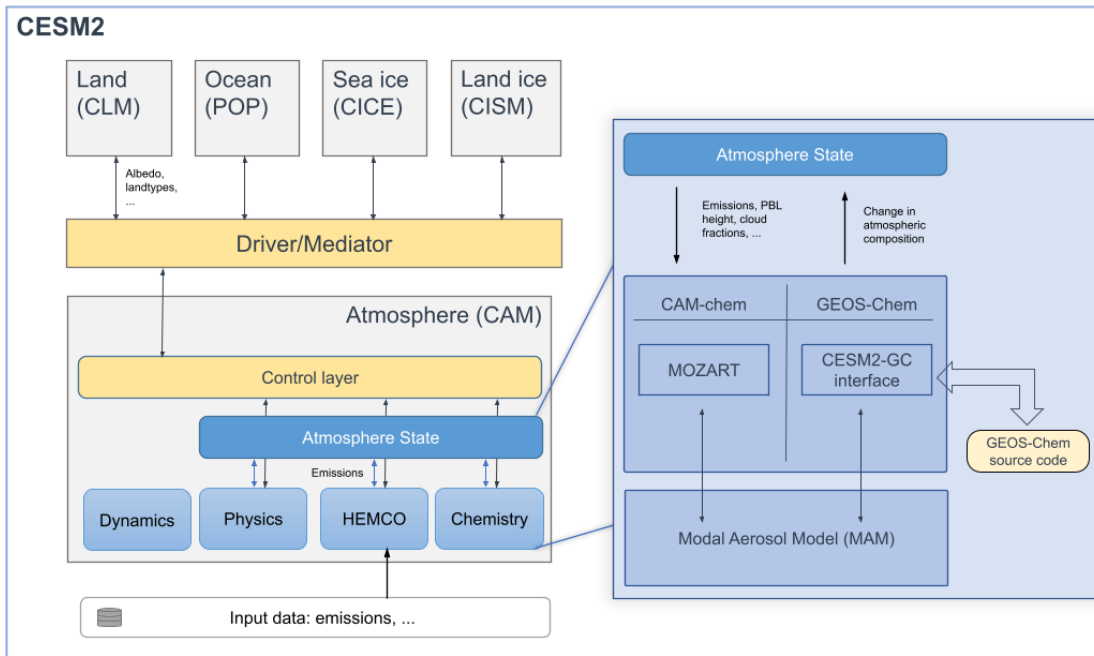



Figure 1. Architectural overview of CESM when running with either the GEOS-Chem or CAM-chem chemistry options. The left section
shows the architecture of CESM, where the five major Earth system components are connected through the driver/mediator. The work
presented here changes only the contents of the atmosphere component (CAM). Regardless of the chemistry option used, dynamics,
physics, and emissions (HEMCO) are handled identically. Each component modifies the "Atmosphere State" while communication occurs
through the control layer. The choice of chemistry module is confined to the "Chemistry" subcomponent, where either CAM-chem or
GEOS-Chem can be chosen. In each case, data are transmitted between the "Atmosphere State" and the chemistry module, which
interacts in turn with the Modal Aerosol Model. Dynamics are shown separately as they act on a "dynamics container" rather than
directly on the atmospheric state. Further detail regarding timing of calls is provided in the Supplementary Information.

At each time step, CESM calls the coupling interface (referred to as CESM2-CG interface in Figure 1) which fills in the meteorological variables required by either CAM-chem or GEOS-Chem. Atmospheric transport and physics are identical whether using CAM-chem or GEOS-Chem to simulate atmospheric chemistry. The interface passes species concentrations from CAM to GEOS-Chem, which are then modified by GEOS-Chem and passed back to CAM. Meteorological data and land data are also passed to GEOS-Chem through the same interface. The routine calls in CAM when using either GEOS-Chem or CAM-chem are identical, with the appropriate chemistry module defined at compilation time such that the calls are routed to the appropriate routines.

The interface handles the conversion of meteorological variables and concentrations of atmospheric constituents between the state variables in CAM and those used in GEOS-Chem. Since GEOS-Chem operates in a "grid-independent" fashion, changes in the grid specification and other upstream modifications to CESM do not necessitate any changes to this interface (Long et al., 2015). Our version of CESM 2.1.1 is modified such that emissions are handled by the Harmonized Emissions Component

(HEMCO), which operates independently of the chemistry module and can provide emissions data to either CAM-chem or GEOS-Chem equally (Lin et al., 2021).

The interface code is kept in a subfolder of chemistry source code (`src/chemistry/geoschem` subfolder), which also contains a copy of the source code for GEOS-Chem. Unlike the implementation of GEOS-Chem within GEOS, we do not use ESMF. However, we plan to develop a NUOPC-based interface as part of future work.

2.2.   Processes represented by CAM-chem and GEOS-Chem

CAM-chem uses the Model for OZone And Related chemical Tracers (MOZART) family of chemical mechanisms to simulate atmospheric chemistry (Emmons et al., 2020). The tropospheric-stratospheric MOZART-TS1 scheme which we demonstrate in our intercomparison involves 186 gas-phase chemical species and includes stratospheric bromine and chlorine chemistry. MOZART-TS1 does not include detailed tropospheric halogen chemistry or short-lived halogen sources such as sea salt bromine, although these will be available in a future release (Badia et al., 2021; Fernandez et al., 2021). Photolysis rates are calculated using a lookup table, based on calculations with the Tropospheric Ultraviolet and Visible (TUV) radiation model (Kinnison et al., 2007). Wet deposition is calculated using the Neu and Prather (2012) scheme for both convective and large-scale precipitation. Dry deposition velocities over land are calculated for each land type by the Community Land Model (CLM) in CESM using the Wesely (1989) resistance scheme with updates described by Emmons et al. (2020). Deposition velocities over the ocean are calculated separately in CAM-chem. Aerosols are represented using the 4-mode Modal Aerosol Model (MAM4), which includes sulfate, black carbon, primary, and secondary organic aerosols (SOAs) (Mills et al., 2016). Ammonium and ammonium nitrate aerosols are calculated with a parameterization using the bulk aerosol scheme (Tilmes et al., 2016). SOAs are simulated using a 5-bin volatility basis set (VBS) scheme, formed from terpenes, isoprene, specific aromatics and lumped alkanes through reaction with OH, ozone and NO_3, with unique yields for each for each combination of volatility and size bin (Tilmes et al., 2019). This more detailed scheme differs from the default MAM SOA scheme that is used in CAM6 (without interactive chemistry). Aerosol deposition, including dry and wet deposition, and gravitational settling (throughout the atmosphere) are calculated in the MAM code of CESM. CAM-chem also uses a volatility basis set (VBS) approach for SOA with five volatility bins, covering saturation concentrations with logarithmic spacing from 0.01 to 100 μg.m^{-3}. CAM-chem explicitly represents Aitken and accumulation mode SOA using two separate tracers for each volatility bin but does not include an explicit representation of non-volatile aerosol.

GEOS-Chem uses a set of chemical mechanisms implemented with the Kinetic PreProcessor (KPP) (Damian et al., 2002). The standard chemical mechanism has evolved continuously from the tropospheric gas-phase scheme described by Bey et al. (2001) and now includes aerosol chemistry (Park, 2004), stratospheric chemistry (Eastham et al., 2014), and a sophisticated tropospheric-stratospheric halogen chemistry scheme (Wang et al., 2019). The scheme present in GEOS-Chem 13.1.2 includes 299 chemical species. Additional "specialty simulations" such as an aerosol-only option and a simulation of the global mercury

215 cycle are present in GEOS-Chem but are not implemented into CESM in this work. Photolysis rates are calculated using the
216 Fast-JX v7 model (Wild et al., 2000). When implemented as the standalone model, wet deposition is calculated for large-scale
217 precipitation using separate approaches for water-soluble aerosols (Liu et al., 2001) and gases (Amos et al., 2012) with
218 calculation of convective scavenging performed inline with convective transport. A different approach is used to simulate wet
219 scavenging for the implementation of GEOS-Chem in CESM (see Section 2.3.4). Dry deposition is calculated using the Wesely
220 (1989) scheme (Wang et al., 1998), but with updates for nitric acid (HNO_3) (Jaeglé et al., 2018), aerosols (Jaeglé et al., 2011;
221 Alexander et al., 2005; Fairlie et al., 2007; Zhang et al., 2001), and over ocean (Pound et al., 2020). The representation of
222 aerosols in GEOS-Chem varies by species. Sulfate-ammonium-nitrate aerosol is represented using a bulk scheme (Park, 2004),
223 with gas-particle partitioning determined using ISORROPIA II (Fountoukis and Nenes, 2007). Modal and sectional size-
224 resolved aerosol schemes are available for GEOS-Chem (Kodros and Pierce, 2017; Yu and Luo, 2009), but are disabled by
225 default and not used in this work. Sea salt aerosol is represented using two (fine and coarse) modes (Jaeglé et al., 2011), while
226 dust is represented using four size bins (Fairlie et al., 2007). We use the "complex SOA" chemistry mechanism in GEOS-
227 Chem when running in CESM, as this uses a volatility basis set (VBS) representation of secondary organic aerosol which is
228 broadly compatible with that used in CAM--chem (Pye and Seinfeld, 2010; Marais et al., 2016; Pye et al., 2010). The complex
229 SOA VBS scheme uses four volatility bins covering saturation concentrations on a logarithmic scale from 0.1 to 100 $\mu g.m^{-3}$.
230 Two classes of SOA are represented in this fashion: those derived from terpenes (TSOA) and those derived from aromatics
231 (ASOA). For each "class" of SOA, two tracers are used to represent each volatility bin (one holding the gas phase mass, the
232 other holding the condensed phase mass). The only exception is the lowest-volatility aromatic aerosol, which is considered to
233 be non-volatile and therefore has no gas-phase tracer. Two additional SOA tracers, representing isoprene-derived and glyoxal-
234 derived SOA, are not represented using a VBS approach.
235
236 Additional differences between the two chemistry modules include the use of different Henry's law coefficients, gravitational
237 settling schemes, representation of polar stratospheric clouds, and heterogeneous chemistry. Full descriptions of the two
238 models are available at https://geos-chem.seas.harvard.edu/ and in Emmons et al. (2020).

2.3.   Representation of atmospheric processes in GEOS-Chem when running in CESM

240 Some processes cannot be easily transferred from standalone GEOS-Chem to its implementation in CESM, due to factors such
241 as the different splitting of convective transport in the two models. Processes which vary in their implementation between the
242 standalone and CESM implementations of GEOS-Chem are described below.

2.3.1.   Aerosol coupling in CESM with GEOS-Chem

244 Since GEOS-Chem and CESM use different approaches to represent aerosols, there is no straight-forward translation between
245 the GEOS-Chem representation and that used elsewhere in CESM. We implement an interface between the CESM and GEOS-

246 Chem representations, so that GEOS-Chem's processing of aerosols is most accurately represented without compromising the

247 microphysical simulations and radiative interactions of aerosol calculated elsewhere in CESM.

248

249 CESM uses the 4-mode version of the Modal Aerosol Model (MAM4) to represent the aerosol size distribution and perform

250 aerosol microphysics (Liu et al., 2016). This represents the mass of sulfate aerosols, secondary organic matter (in five volatility

251 basis set bins), primary organic matter, black carbon, soil dust, and sea salt with advected tracers for each mode (accumulation,

252 Aitken, coarse, and primary carbon), although some species are considered only in a subset of the four modes. A tracer is also

253 implemented for the number of aerosol particles in each mode, resulting in a total of 18 tracers. As discussed above, GEOS-

254 Chem instead represents sulfate, nitrate, and ammonium aerosol constituents with three tracers; fresh and aged black and

255 organic carbon with four tracers; fine and coarse sea salt as two tracers; and different sizes of dust with four tracers. Six

256 additional tracers are used to track the bromine, iodine, and chlorine content of each mode of sea salt aerosol, with two more

257 used to track overall alkalinity. Gas-phase sulfuric acid (H_2SO_4) is assumed to be negligible in the troposphere and is estimated

258 using an equilibrium calculation in the stratosphere (Eastham et al., 2014). The GEOS-Chem mechanism therefore represents

259 greater chemical complexity but reduced size resolution compared to the aerosol representation in MAM4.

260

261 Accordingly, when receiving species concentrations from CESM, the interface to GEOS-Chem lumps all modes of the MAM

262 aerosol into the corresponding GEOS-Chem tracer. This includes gas-phase H_2SO_4, in the case of the GEOS-Chem sulfate

263 (SO_4) tracer. Aerosol constituents which are not represented explicitly by MAM (e.g. nitrates) are not included in this

264 calculation. The relative contribution of each mode is stored during this "lumping" process for each grid cell. Once calculations

265 with GEOS-Chem are complete, the updated concentration of the lumped aerosol is repartitioned into the MAM tracers based

266 on the stored relative contributions in each grid cell.

267

268 For SOAs, additional steps are needed. For the bins covering saturation concentrations of 1 µg.m^{-3} and greater, we assume that

269 the relevant volatility bin in MAM4 is equal to the sum of the two classes in GEOS-Chem covering the same saturation

270 concentrations. For example, the tracers TSOA1 and ASOA1 in GEOS-Chem are combined to estimate the total quantity of

271 the Aitken and accumulation modes for species "soa3" in MAM4. Partitioning between the two modes (when transferring from

272 GEOS-Chem to MAM4) is calculated based on the relative contribution of each constituent to the total prior to processing by

273 GEOS-Chem. Partitioning between the two classes (when transferring from MAM4 to GEOS-Chem) is calculated based on

274 the relative contribution of each constituent to the total at the end of the previous time step. For the lowest-volatility species,

275 we split the lowest volatility bin concentrations (and non-volatile species) from GEOS-Chem between the two lowest

276 volatilities in MAM4. A full mapping for all species is provided in Table 1.

277

278 *Table 1. Mapping between tracers used to represent SOA in GEOS-Chem and CAM-chem. Translation between GEOS-Chem and MAM4 is*
279 *performed by preserving the relative contributions provided during the previous transfer.*

GEOS-Chem species	Mapping to CAM-chem species	Saturation concentration range ($\mu g.m^{-3}$)	Phase
TSOA0 + ASOAN	soa1_a1 + soa1_a2 + soa2_a1 + soa2_a2	0 – 0.1	Aerosol
TSOA1 + ASOA1	soa3_a1 + soa3_a2	0.1 – 1.0	Aerosol
TSOA2 + ASOA2	soa4_a1 + soa4_a2	1.0 – 10	Aerosol
TSOA3 + ASOA3	soa5_a1 + soa5_a2	10 – 100	Aerosol
TSOG0	SOAG0 + SOAG1	0 – 0.1	Gas
TSOG1 + ASOG1	SOAG2	0.1 – 1.0	Gas
TSOG2 + ASOG2	SOAG3	1.0 – 10	Gas
TSOG3 + ASOG3	SOAG4	10 – 100	Gas

280

281 Finally, MAM simulates some chemical processing on and in the aerosol. This includes the reaction of sulfur dioxide with
282 hydrogen peroxide and ozone in clouds, which is already included in the GEOS-Chem chemistry mechanism. We therefore
283 disable in-cloud sulfur oxidation in MAM4 when using the GEOS-Chem chemistry component in CESM, consistent with the
284 GEOS-Chem CTM. A comparison of the effect of each approach is provided in the Supplementary Information.

285 **2.3.2.   Dry deposition**

286 Dry deposition velocities over land are calculated in CESM for each atmospheric constituent by the Community Land Model
287 (CLM) using a species database stored by the coupler. GEOS-Chem is also able to calculate its own dry deposition velocities
288 (see Section 2.2), in situations where a land model is not available such as when running as a CTM. We thus implement
289 different options to compute dry deposition velocities when running CESM with the GEOS-Chem chemistry option:

290   1. Dry deposition velocities over land are computed by CLM and are passed to CAM through the coupler. They are then
291   merged with dry deposition velocities computed over ocean and ice by GEOS-Chem, identical to the procedure used
292   in CAM-chem. Each of these are scaled by the land and ocean/ice fraction respectively.
293   2. GEOS-Chem computes dry deposition at any location using the land types and leaf area indices from CLM, which
294   are passed through the coupler.
295   3. GEOS-Chem obtains "offline" land types and leaf area indices and computes the dry deposition velocities similarly
296   to GEOS-Chem Classic.

297 This allows researchers to experiment with different dry deposition options, ranging from that most consistent with the
298 approach used in CAM-chem (option 1) to that most consistent with stand-alone GEOS-Chem (option 3). For this work we
299 use option 2, but option 1 will be brought as standard into the CESM main code to reduce data transfer requirements.

2.3.3. Emissions

The Harmonized Emissions Component (HEMCO) is used to calculate emissions in standalone GEOS-Chem (Keller et al., 2014), and HEMCO v3.0 was recently implemented as an option for CAM-chem (Lin et al., 2021). HEMCO offers the possibility for the user to read, regrid, overlay, and scale emission fluxes from different archived emissions inventories at runtime. Emissions extensions allow for the computation of emissions that depend on meteorology or surface characteristics (e.g. lightning, dust emissions). Some extensions have also been designed to calculate subgrid-scale chemical processes, such as non-linear chemistry in ship plumes (Vinken et al., 2011).

The GEOS-Chem CTM implementations use archived ("offline") inventories of natural emissions, calculated at native resolution using the NASA GEOS MERRA-2 and GEOS-FP meteorological fields. This ensures that the emissions are calculated consistently regardless of grid resolution. These archived emissions fields can be used within CESM but we also preserve the option for users to employ "online" emissions inventories where relevant. This enables feedback between climate and emissions to be calculated. For instance, lightning nitrogen oxides ($NO_x = NO + NO_2$) emissions, dust and sea salt emissions, and biogenic emissions are all computed online using parameterizations from CAM and CLM. CAM computes lightning NO_x emissions based on the lightning flash frequency, which is estimated following the model cloud height, with different parameterizations over ocean and land. The NO lightning production rate in CAM is assumed proportional to the discharged energy, with 10^{17} atoms of nitrogen released per Joule (Price et al., 1997). The lightning NO_x emissions are then allocated vertically from the surface to the local cloud top based on the distribution described by Pickering et al. (1998). For biogenic emissions, we use the online Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGANv2.1), as established in CLM (Guenther et al., 2012). Aerosol mass and number emissions are passed directly to MAM constituents. Global anthropogenic emissions can be specified from any of the standard GEOS-Chem inventories, but default to the Community Emissions Data System (CEDS) inventory (Hoesly et al., 2018). Sulfur emissions from the CEDS inventory are partitioned into size-resolved aerosol (mass and number) and SO_2 (Emmons et al., 2020). In CAM, volcanic out-gassing of SO_2 is provided from the GEIA inventory with 2.5% emitted as sulfate aerosol (Andres and Kasgnoc, 1998), while eruptive emissions are provided from the VolcanEESM database (Neely and Schmidt, 2016). The option is also available through HEMCO to use the "AeroCom" volcanic emissions, which are derived from OMI observations of SO_2 (Ge et al., 2016; Carn et al., 2015).

Although we use HEMCO with both model configurations, there remain differences between the representation of emissions in CAM-chem and in GEOS-Chem when run within CESM. This is because of differences in the species present in their respective mechanisms. For instance, emissions of iodocarbons (CH_3I, CH_2I_2, CH_2ICl, CH_2IBr) and inorganic iodine (HOI, I_2) are not available in CAM-chem since iodine is not explicitly modeled in the versions of CAM-chem available in CESM v2.1.1. VOC lumping is also performed differently (see the Supplemental Information for more detail).

333

Where the emitted species are present in both chemical mechanisms, the emissions calculated by HEMCO in CESM are identical whether running with GEOS-Chem or CAM-chem. If the HEMCO implementations of lightning, dust, sea salt, and biogenic emissions are used, emissions will be identical between CESM and the standalone GEOS-Chem CTM.

2.3.4.   Wet deposition and convection

For both GEOS-Chem and CAM-chem within CESM, convective scavenging and transport are handled separately. Unlike in the Liu et al. (2001) approach implemented in the GEOS-Chem standalone code, removal of soluble gases within convective updrafts is not explicitly simulated in either CAM-chem or GEOS-Chem when embedded in CESM. When using the CAM-chem mechanism within CESM, the Neu scheme is used to perform washout of soluble gaseous species, while wet deposition of MAM aerosols is handled by MAM. When running CESM with the GEOS-Chem chemistry mechanism, the Neu scheme also performs wet scavenging for aerosols which are not represented by MAM4 (e.g. nitrate). For all such aerosols we assume a Henry's law coefficient equal to that for HNO_3.

2.3.5.   Surface boundary conditions

In CESM, surface boundary mixing ratios of long-lived greenhouse gases (methane, N_2O, and chlorofluorocarbons) are set to the fields specified for CMIP6 historical conditions and future scenarios (Meinshausen et al. 2017). For whichever CMIP6 scenario is chosen, the boundary conditions overwrite those set by the GEOS-Chem chemistry module or by the HEMCO emissions component.

2.4.   Changes to the data flow in CESM when running with GEOS-Chem

In CESM, data such as the Henry's law coefficients required to calculate dry deposition velocities and wet scavenging rates for each species are defined at compile time. For species that are common to GEOS-Chem and CAM-chem but where these factors differ, the GEOS-chem values are used by default. The CAM-Chem values are listed alongside them in the source code to allow users to switch if desired. Additionally, we modify CAM, CLM and the Common Infrastructure for Modeling the Earth (CIME) such that the land model can pass land type information and leaf area indices to the atmosphere model to compute dry deposition velocities. This could be a potential solution for dry deposition of aerosols in MAM, which currently uses fixed land types independent of the ones used in CLM (Liu et al., 2012). However, this comes at the cost of passing land information through the coupler at every time step.

2.5.   Installation and compilation process

The interface between CESM and GEOS-Chem, as well as the GEOS-Chem source code, is automatically downloaded when CAM checks out its external repositories. The versions of GEOS-Chem and of the coupling interface can be changed by modifying the `Externals_CAM.cfg` and by running the `checkout_externals` command.

363

364 When creating a new case, the user chooses the atmospheric chemistry mechanism (GEOS-Chem or CAM-chem). The

365 chemistry option is defined by the name of the CESM configuration (component set, or "compset"), making the process of

366 creating a run directory almost identical when choosing either GEOS-Chem or CAM-chem. Whereas chemistry options in

367 CAM-chem are set explicitly using namelist files, certain options in GEOS-Chem are set using ASCII text input files which

368 are read during the initialization sequence. The installation and build infrastructure of CIME will therefore copy any GEOS-

369 Chem specific text input files to the case directory when setting up a simulation which includes GEOS-Chem. This currently

370 includes emissions specifications read by HEMCO, although this is expected to change as HEMCO becomes the standard

371 emissions option for both CAM-chem and GEOS-Chem in CESM (currently being discussed with the CESM team).

372

373 Although CESM supports both shared-memory parallelization (OpenMP) and distributed memory parallelization (MPI),

374 GEOS-Chem implemented in CESM does not currently support OpenMP. When running CESM with the GEOS-Chem

375 chemistry model, the number of OpenMP threads per MPI task is therefore set to one.

376

377 Although a complete copy of the GEOS-Chem source code is downloaded from the version-controlled remote of GEOS-Chem

378 repository (to ensure that the most-recent release of GEOS-Chem is used), not all files present in the GEOS-Chem source code

379 directory are compiled. For instance, the files pertaining to the GEOS-Chem advection scheme are not needed as advection is

380 performed by CAM, and therefore the GEOS-Chem advection routines are not compiled. To do this we implement a new

381 feature in CIME to use `.exclude` files which list files not needed during compilation. CIME reads each `.exclude` file at

382 compile time and searches subdirectories recursively from the location of the exclude file, preventing any named file from

383 being included in compilation. For example, an `.exclude` file is provided in the chemistry coupling interface folder for

384 GEOS-Chem that lists the files to exclude in the GEOS-Chem source code directories.

385 **3.   Setup used for model evaluation**

386 We simulate a two-year period with GEOS-Chem embedded in CESM (hereafter C-GC). The simulation setup is described in

387 the present Section. Then, we perform a comparison of its output to that generated by two other model configurations (Section

388 4). By comparing the results to those produced for the same period by CESM with CAM-chem (hereafter C-CC), we can

389 perform the first comparison of GEOS-Chem and CAM-chem when run as chemistry modules within the same ESM. Any

390 differences between these two simulations can only be the result of differences between the two chemical modules and their

391 implementations in CESM. This includes not only differences in the gas-phase chemical mechanism, but also in the

392 implementation of photolysis calculations, heterogeneous chemistry, aerosol microphysics, and the chemical kinetics

393 integrator itself. We also compare output to that produced by the standalone GEOS-Chem High Performance model (hereafter

S-GC). This enables us to evaluate the effect of using CESM's grid discretization, advection, aerosols, and representation of meteorology compared to that used in the GEOS-Chem CTM.

Lastly, we evaluate the performance of C-GC by comparing output to observational data (Section 5). We also include comparisons of data from the C-CC and S-GC configurations, to provide insight into the relative performance of the model and the root cause of disagreements with observations. This section describes the model configurations in detail, but a brief summary is provided in Table 6.

All simulations cover January 1st to December 31st 2016, with an additional 6 months (S-GC) or 1 year (C-GC/C-CC) of spin up. For C-CC, the standard restart file provided with CESM is used to provide initial conditions. For S-GC, we use a restart file provided with version 13.1.2 of the GEOS-Chem chemistry module, which was obtained from a 10-year simulation. The CESM restart file is intended to represent the early 21st century, so we have followed the lead of previous studies which have used a 1-2 year spin up period (Schwantes et al., 2022; He et al., 2015). For C-GC, we use initial conditions which are taken from the S-GC restart file where possible, but fill missing species (e.g. MAM4 aerosol tracers) using data from the C-CC restart file. Both simulations performed with CESM v2.1.1 (C-GC and C-CC) use a horizontal resolution of 1.9°×2.5° on 56 hybrid pressure levels, extending from the surface to 1.65 hPa. Aerosols are represented in CESM using the 4-mode version of the modal aerosol model, MAM4 (Liu et al., 2012). In C-GC, we use the complex SOA chemistry scheme (Pye and Seinfeld, 2010; Pye et al., 2010; Marais et al., 2016). In C-CC, we use the MOZART-TS1 chemistry scheme (Emmons et al., 2020).

Standalone GEOS-Chem (S-GC) simulations are performed using the GEOS-Chem High Performance (GCHP) configuration, using a C48 cubed-sphere grid (approximately equivalent to a 2°×2.5° horizontal grid) on 72 hybrid pressure levels extending up to 0.01 hPa. In GCHP, chemistry is performed up to 1 hPa (approximately 50 km) with simplified parameterizations used above that point. Aerosols are represented using GEOS-Chem's "native" scheme, without translation to or from MAM4. As in C-GC, we use the complex SOA scheme.

All three model configurations are driven using meteorological data from MERRA-2. In S-GC all meteorological fields are explicitly specified by MERRA-2, using the same 72-layer vertical grid. The only exception is the specific humidity in the stratosphere, which is computed online. In C-CC and C-GC, we use the "specified dynamics" (SD) configuration of CAM6 in which 3-D temperature, 3-D wind velocities, surface pressure, surface temperature, surface sensible heat flux, surface latent heat flux, surface water flux, and surface stresses are provided by MERRA-2 on a truncated 56-layer vertical grid. These variables are nudged with a relaxation time of 50 hours, resulting in a relatively "loose" nudging strength. All other fields (e.g. cloud fraction) are computed using the CAM physics routines. This includes convection. Whereas S-GC computes convective transport from archived convective mass fluxes and calculates scavenging within the updraft (Wu et al., 2007), convective transport in both C-CC and C-GC is calculated in CAM6 using the CLUBB-SGS scheme for shallow convection (Bogenschutz

428 et al., 2013) and the Zhang-McFarlane scheme (Zhang and McFarlane, 1995) for deep convection. Scavenging within the
429 convective updraft is not simulated explicitly.
430
431 Water vapor in C-GC is initialized from the specific humidity "Q" restart variable, which is identical to the one used for C-
432 CC; after this point humidity is calculated based on the moist processes represented explicitly in CAM's physics package. The
433 GEOS-Chem CTM does not calculate water vapor in the troposphere, instead prescribing specific humidity directly from
434 MERRA-2 output. Mixing ratios of water vapor in C-CC and C-GC are therefore identical to that in S-GC at initialization
435 time, but from that point onwards may diverge.
436
437 Emissions are harmonized between the three models, with all three configurations using HEMCO to calculate emissions fluxes.
438 Surface anthropogenic emissions are provided from CEDS and are identical between all three models, apart from small
439 differences in effective emissions from ships due to parameterized plume processing (Vinken et al., 2011). Simulated
440 anthropogenic and biomass burning surface emissions of nitrogen oxides are 128-132 Tg(N) in each of the three models.
441 Aviation emissions are calculated in all three models based on the AEIC 2005 emission inventory, contributing a further
442 0.8 Tg(N) in addition to other species (Simone et al., 2013).
443
444 Lightning emissions are calculated in C-CC and C-GC using the online parameterization described in Section 2.3.3, while
445 lightning emissions in S-GC are calculated using archived flash densities and cloud top heights (Murray et al., 2012). Total
446 lightning NO_x emissions are 5.7-6.1 Tg(N) in all three models. A summary of the breakdown of NO_x emissions is provided in
447 Table 2.
448
449 *Table 2. Annual global anthropogenic, soil, and lightning NO_x emissions expressed in Tg(N)/year.*

	C-GC	**S-GC**	**C-CC**
Anthropogenic + biomass burning	40.1	39.1	39.6
Soil	7.23	7.23	7.23
Lightning	6.05	5.82	5.71

450
451 Biogenic emissions are calculated in C-CC and C-GC using the embedded MEGAN emissions module in CESM, which differs
452 slightly from the implementation in S-GC and will produce different emissions due to different vegetation distributions. Total
453 biogenic emissions in S-GC and C-GC are shown in Table 3. In all three simulations we use the "AeroCom" volcano emissions
454 implemented in HEMCO.
455

456 *Table 3. Annual global biogenic emission totals in GEOS-Chem implemented in CESM (C-GC) compared to in standalone GEOS-Chem (S-*
457 *GC).*

Species	Name in GEOS-Chem	C-GC (Tg/year)	S-GC (Tg/year)
Acetone	ACET	42.7	48.2
Acetic acid	ACTA	3.86	-
Acetaldehyde	ALD2	20.8	17.9
Lumped alkanes >= C_4	ALK4	0.16	-
Ethylene	C2H4	30.4	-
Ethane	C2H6	0.34	0.21
Propane	C3H8	0.03	-
Formaldehyde	CH2O	5.14	-
Carbon monoxide	CO	88.8	-
Ethanol	EOH	20.8	17.9
Limonene	LIMO	11.0	9.11
α/β-pinene, sabinene, carene	MTPA	98.6	81.5
Other monoterpenes	MTPO	40.8	38.6
Isoprene	ISOP	502	397.6
Methanol	MOH	119	-
Toluene	TOLU	1.57	-
Lumped alkenes >= C_3	PRPE	22.3	24.2

458

459 Emissions of aerosols (primary organic matter, and black carbon) are listed in Table 4. These emissions are consistent with the

460 values provided in Tilmes et al. (2016).

461

462 *Table 4. Annual global emissions of sulfates, primary organic matter and black carbon in all three model configurations.*

	C-GC	S-GC	C-CC
SO_4 in Tg(S)/year	0.22	0.22	0.22
POM in Tg(C)/year	45.86	34.57	57.19
BC in Tg(C)/year	8.14	7.86	6.24

463

464 Mobilization of mineral dust is calculated in all three models using the DEAD scheme (Zender, 2003). In C-CC and C-GC,

465 the online implementation in CESM is employed, resulting in total natural mineral dust emissions of 5984 Tg/year. A brief

466 discussion of dust emissions in CESM is provided in the Supplementary Information. In S-GC, natural mineral dust emissions

467 are calculated online using the same scheme but with a different scaling and at a slightly different grid resolution, resulting in

468 total emissions of 1390 Tg/year.

469

470 Emissions of sea salt are calculated online in CESM for C-GC and C-CC, while S-GC uses a pre-calculated (offline) inventory

471 of sea-salt emissions, as well as sea-salt bromine and chloride. Emissions of sea-salt bromine in C-GC are calculated based on

472 the offline inventory rather than the calculated emissions of sea salt, and therefore do not scale correctly with the estimated

473 sea-salt emissions from CESM (see Table 5). This will be resolved as part of future work.

474

475 *Table 5. Annual global emissions of sea salt aerosols (fine and coarse) and bromine in sea salt for C-GC and S-GC. The names of the tracers*
476 *used to represent these species in GEOS-Chem are provided in brackets.*

Species	C-GC (Tg/year)	S-GC (Tg/year)
Fine sea-salt (SALA)	93.0	59.1
Coarse sea-salt (SALC)	2780	3576
Bromine in fine sea-salt (BrSALA)	0.166	0.126
Bromine in coarse sea-salt (BrSALC)	10.1	7.54

477

478 Finally, for long-lived species such as chlorofluorocarbons (CFCs) we use the shared socio-economic pathway 2-4.5 (SSP2-

479 4.5) (Riahi et al., 2017) set of surface boundary conditions in both C-GC and C-CC. In comparisons against S-GC we use

480 historical emissions from the World Meteorological Organization's 2018 assessment of ozone depletion (Fahey et al., 2018).

481 However, this difference is unlikely to significantly affect simulation output given the short duration of the simulations.

482

483 *Table 6. Brief summary of the model configuration used for C-GC, S-GC, and C-CC.*

	C-GC	S-GC	C-CC
Horizontal resolution	1.9°×2.5°	C48 (~2°×2.5°)	1.9°×2.5°
Vertical levels	L56 (up to 1.65 hPa)	L72 (up to 0.01 hPa)	L56 (up to 1.65 hPa)
Aerosol microphysics	MAM4 with VBS-SOA	Bulk with VBS-SOA	MAM4 with VBS-SOA
Aerosol tracers	Mixed (S-GC for aerosol chemistry; C-CC for microphysics)	SO4, BC, OM, sea salt, dust, AERI, DMS, INDIOL, IONITA, MONITA, MOPI, MOPO, MSA, NH4, NIT, pFe, SOAGX, SOAIE, TSOA, ASOA	SO4, BC, OM (both primary and secondary), sea salt, dust
Treatment of SOA	Explicit calculation of SOA using VBS (five bins)	Explicit calculation of SOA using VBS (four bins)	Explicit calculation of SOA using VBS (five bins)
Chemistry	GEOS-Chem 13.1.2	GEOS-Chem 13.1.2	MOZART-TS1
Biogenic emissions	Online from dynamically evolving vegetation computed in CLM using MEGAN2.1	Offline from archived vegetations using MEGAN2.1	Online from dynamically evolving vegetation computed in CLM using MEGAN2.1

| Original model references | This work | (Bey et al., 2001) | (Tilmes et al., 2016; Emmons et al., 2020; Lamarque et al., 2012) |

4. Model intercomparison

We first compare the global distribution of ozone and aerosols between C-GC, S-GC, and C-CC. Section 4.1 evaluates the vertical and latitudinal distribution of ozone and two related species (water vapor (H_2O) and the hydroxyl radical (OH)), followed by the global distribution of ozone at the surface in each model configuration (Section 4.2). Stratospheric chemistry in GEOS-Chem is described by Eastham et al. (2014) and by Emmons et al. (2020) for CAM-chem. A similar evaluation of differences in zonal mean and surface aerosol concentrations follows (Section 4.3).

To understand the causes of these differences, we compare the global distribution of reactive nitrogen and halogen species in each model configuration (Section 4.4). When comparing halogen distributions we consider only bromine and chlorine distributions, as iodine is not simulated in this version of CAM-chem. The latest implementation of halogen chemistry in GEOS-Chem and its role in atmospheric chemistry are described by Wang et al. (2021), while its representation in CAM-chem is described by Emmons et al. (2020). Differences in the total atmospheric burden and vertical distribution of these families provides information regarding differences in removal processes. Differences in their internal partitioning (e.g. between NO_x and HNO_3) provide information regarding the representation of atmospheric chemistry.

4.1. Ozone

Figure 2 shows the annual mean mixing ratio of stratospheric ozone simulated by each of the three model configurations. At 10 hPa in the tropics, where ozone mixing ratios reach their peak, the three configurations agree to within 10% suggesting a reasonable representation of stratospheric ozone. However, near the tropopause the three configurations diverge. Comparison of C-GC to S-GC (panel b) shows mixing ratios 20% lower near the tropical tropopause but more than 50% greater in the extratropical lower stratosphere. However, C-GC simulates mixing ratios of ozone around the tropopause which are 20% lower than C-CC (panel c) at all latitudes.

505

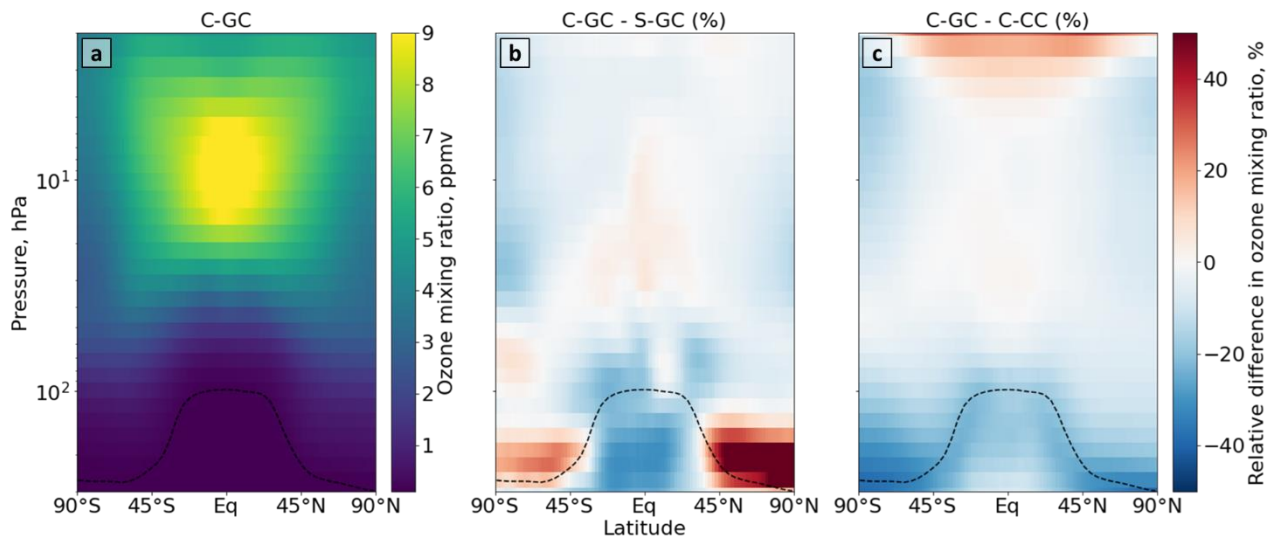



Figure 2. Comparison of stratospheric ozone simulated with CESM running GEOS-Chem (C-GC) to standalone GEOS-Chem (S-GC) and CESM running CAM-chem (C-CC). Left column (a): absolute values estimated with C-GC. Center column (b): relative difference between C-GC and S-GC. Right column (c): relative difference between C-GC and C-CC. Red (blue) shading means that C-GC estimated a higher (lower) value than the other model.

510 The difference in pattern in the comparison between C-GC, S-GC, and C-CC implies that the cause is likely to be related to

511 factors which are common between C-GC and C-CC, such as the representation of meteorology. One such factor may be water

512 vapor, which is treated differently between the "online" (C-GC, C-CC) and "offline" (S-GC) configurations.

513

514 To quantify and understand these differences in stratospheric ozone, we analyze concentrations of three different related

515 compounds from the surface to the stratosphere: ozone, the hydroxyl radical OH, and water vapor. OH reacts with most trace

516 species in the atmosphere and its high reactivity makes it the primary oxidizing species in the troposphere, such that differences

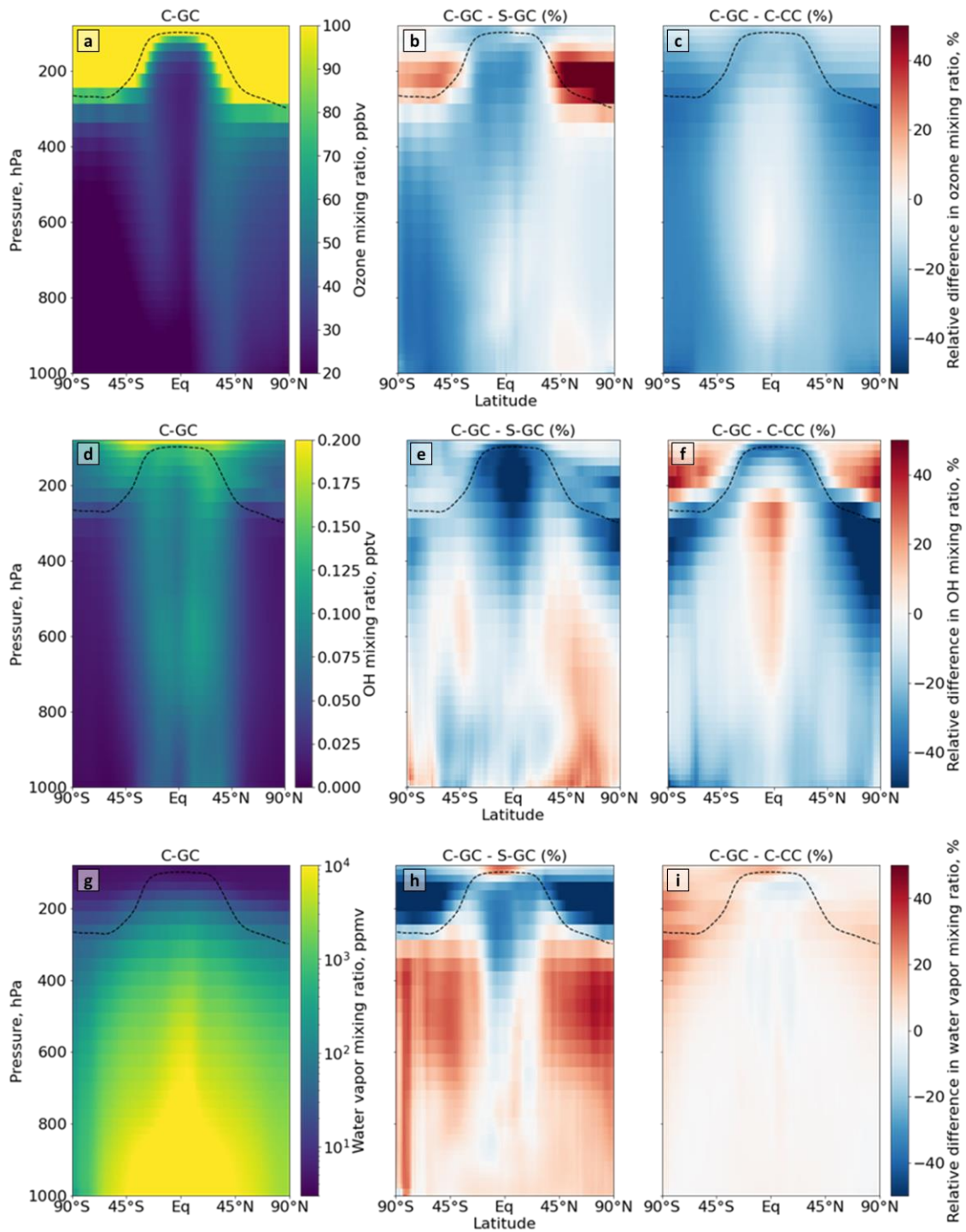
517 in abundance between models will affect the simulated abundances of many atmospheric pollutants (Seinfeld and Pandis,

518 2006). Since OH is produced from water vapor and (indirectly) ozone, these three compounds can collectively be used to

519 understand some of the differences between C-GC, S-GC, and C-CC. Later analyses will focus on NO_x, bromine, and chlorine,

520 each of which also strongly affect tropospheric and stratospheric concentrations of ozone.

521

522

523   *Figure 3. Comparison of atmospheric composition simulated with CESM running GEOS-Chem (C-GC) to standalone GEOS-Chem (S-GC)*
524   *and CESM running CAM-chem (C-CC). Different rows show different constituents, while different columns show different model results.*
525   *Top row (a-c): ozone. Middle row (d-f): OH radical. Bottom row (g-i): water vapor. Left column (a, d, g): absolute values estimated with*

528 The upper row of Figure 3 shows the distribution of ozone as represented by C-GC (panel a), and the difference when compared

529 to S-GC (panel b) or C-CC (panel c). Comparing to C-CC, C-GC estimates mixing ratios of ozone which are 30% lower from

530 the surface (across all latitudes) and throughout the extratropical troposphere. This is consistent with previous work which

531 showed that ozone simulated by GEOS-Chem to match the KORUS-AQ campaign had a normalized mean bias of -26%,

532 compared to -9% in CAM-chem (Park et al., 2021). In the present study, we find that ozone mixing ratios around the tropopause

533 are also lower in C-GC than in C-CC by 15-20%. This suggests that discrepancies observed in KORUS-AQ may be related to

534 chemistry rather than the treatment of meteorology, but a more focused regional analysis would be needed to confirm this.

535

536 Comparing to the differences between C-GC and S-GC provides some insight into possible causes for these discrepancies.

537 Near-surface ozone in C-GC in the Southern Hemisphere is also 30-40% lower than in S-GC, suggesting a potential common

538 cause for the differences with C-CC. However, in the Northern extratropical troposphere below 400 hPa, zonal mean

539 differences between C-GC and S-GC are consistently less than 10%. Ozone concentrations are also lower in the tropical mid-

540 troposphere in C-GC than in S-GC by 15-25%, whereas concentrations were well matched in this region between C-GC and

541 C-CC. In the lower stratosphere, ozone concentrations in C-GC are instead greater than in S-GC, with the difference in the

542 Northern extratropical lower stratosphere exceeding 50%. The global ozone burden in C-GC is within 1.5% of that estimated

543 by S-GC, while C-CC has a total atmospheric ozone burden 15% greater than C-GC. These model differences are evaluated

544 against observations in Section 5.2.

545

546 Differences in tropospheric NO_y and halogens, in particular the higher loading of BrO in C-GC, may explain some of these

547 differences (see Section 4.4). However, another possible factor in these differences in ozone is differences in water vapor

548 distribution. The bottom row of Figure 3 shows the annual average simulated distribution of water vapor in C-GC, and the

549 difference relative to S-GC and C-CC. Water vapor concentrations are approximately equal between C-GC and C-CC, since

550 the representation of moist physics in the two models is identical. However, differences of up to 20% arise around the

551 tropopause, possibly due in part to the different representation of stratospheric water chemistry and settling of stratospheric

552 aerosol (including ice). This is unlikely to be due to HO_x catalytic cycles depleting ozone, as OH in this region is lower in C-

553 GC than in S-GC (panel e) and HO_x cycles are in any case a minor contributor to ozone depletion in the lower stratosphere

554 (Brasseur and Solomon, 2006). The greater water vapor (and therefore humidity) may instead result in faster heterogeneous

555 chemistry, including the liberation of NO_x from HNO_3.

556

557 The differences between C-GC and S-GC are larger. Outside of the tropics and below the tropopause, water vapor

558 concentrations are up to 30% greater in C-GC than in S-GC. Differences are smaller in the tropics, but in the tropical upper

559 troposphere water vapor concentrations are instead 15% lower in C-GC than in S-GC. This may be part of the reason that

water vapor concentrations in the extratropical lower stratosphere are more than 50% lower in C-GC than in S-GC, since the tropical upper troposphere is the source of water vapor to the stratosphere. This is the same region in which C-GC calculates ozone mixing ratios which are more than 50% greater than in S-GC, potentially due to the lower concentration of water vapor (an indirect sink for ozone).

These differences arise due to the different representation of moist processes between CAM's physics package (used in both C-GC and C-CC), and GEOS, which produces MERRA-2 and therefore is represented in S-GC. For example, although total annual average precipitation agrees to within 10% between the models, the mean volumetric cloud fraction in C-GC and C-CC is 15%, compared to 8% in S-GC. Meanwhile the area-averaged cloud water content and cloud ice content are 57% and 38% greater in S-GC than in C-GC (or C-CC).

Differences in ozone and water vapor result in differences in concentrations of OH, as shown in the middle row of Figure 3. The global OH atmospheric burden is approximately 10% lower in C-GC than in S-GC, but this difference is not evenly distributed. Differences in OH concentrations can be roughly considered to be the product of differences in ozone and differences in water vapor, since both are needed to create OH (along with UV radiation). In the tropical troposphere, OH concentrations are more than 50% lower in C-GC than in S-GC, likely due to a relative lack of both ozone and water vapor. However, in the Northern mid- and upper latitudes below 900 hPa, OH concentrations are 10-20% greater in C-GC than in S-GC. This reflects the greater water vapor concentrations and roughly equal ozone concentrations between the two models.

4.2. Surface ozone

Figure 4 compares the simulated, annually-averaged surface ozone mixing ratios as estimated by C-GC, S-GC, and C-CC. We find that, when globally averaged, C-GC predicts a lower surface ozone mixing ratio than either C-CC or S-GC. Averaged over each Hemisphere, C-GC estimates a lower surface ozone mixing ratio than S-GC (panel b) by 4.9 ppbv and 2.2 ppbv in the Southern Hemisphere and Northern Hemisphere respectively. This varies between the land and oceans. In the Northern Hemisphere, we observe a small difference in surface ozone mixing ratio over the oceans (less than 1 ppbv), while a difference of approximately 3 ppbv can be found over North America, Europe and East Asia.

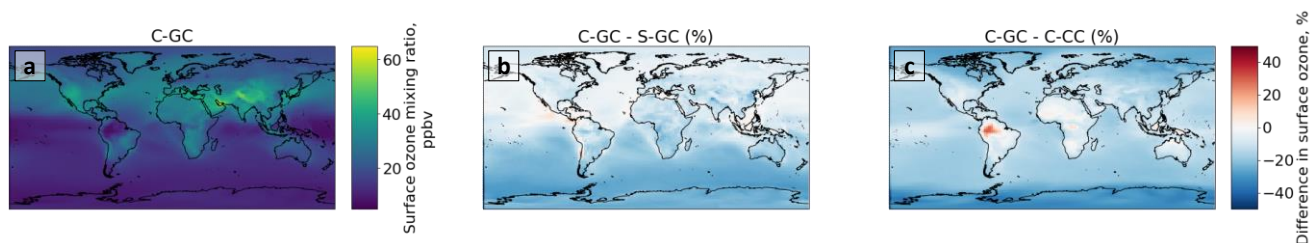



Figure 4. Comparison of the annually averaged surface ozone mixing ratios simulated with CESM running GEOS-Chem (C-GC, panel a) to standalone GEOS-Chem (S-GC, panel b) and CESM running CAM-chem (C-CC, panel c). Red (blue) shading means that C-GC estimated a higher (lower) value than the other model.

The difference between C-GC and C-CC (panel c) does not show the same hemispheric asymmetry in absolute terms, and a larger difference over oceans than over land. We find that C-GC estimates 5.4 and 7.9 ppbv less ozone than C-CC in the Southern and Northern Hemispheres respectively. The pattern indicated in Figure 4c suggests that bromine from sea salt may be the principal cause of the differences in surface ozone between C-GC and C-CC, whereas differences between C-GC and S-GC are likely to be related to anthropogenic emissions given the hemispheric asymmetry. The 20-30% increase in ozone over the Amazon in C-GC related to C-CC may instead be related to differences in biogenic emissions.

In addition to annual averages, we also consider seasonal variations of surface ozone. Figure 5 presents parity plots of monthly-averaged surface ozone mixing ratios for January and July comparing C-GC to S-GC and C-CC, after outputs from all three model configurations were remapped to a common 2°×2.5° grid. In January, we find a correlation coefficient of 0.91 and slope of 0.91 between C-GC and S-GC. In July this agreement is worsened, with a correlation coefficient of 0.80 but a slope of 0.93. This indicates that the sources of differences in surface ozone mixing ratios between C-GC and S-GC are magnified during boreal summer. There is also a distinctive "hot spot" in the July parity plot, with a large cluster of grid cells showing mixing ratios in the range 20-25 ppbv in S-GC but 10-20 ppbv in C-GC. Further research is needed to establish the origin of this cluster, which does not occur during boreal winter.

607

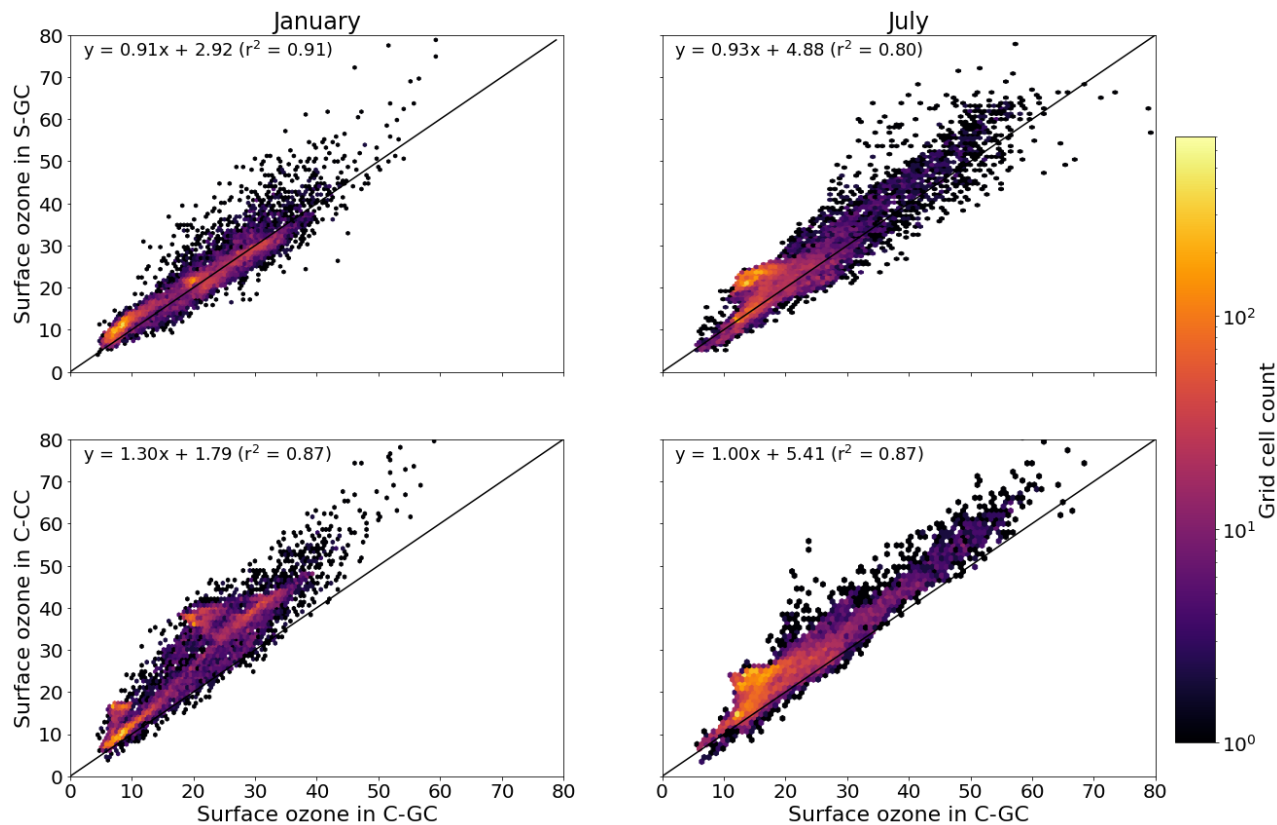
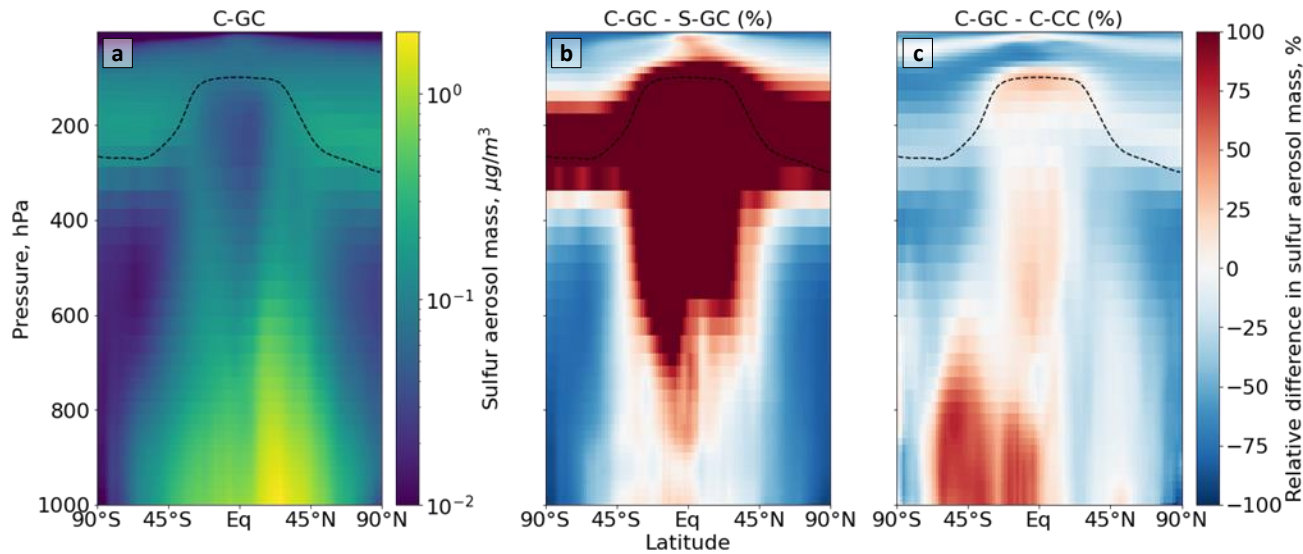



Figure 5. Parity plots of surface ozone mixing ratios, expressed in ppbv, for January (left) and July (right) comparing C-GC on the X axis to S-GC (top) and C-CC (bottom) on the Y axis. Fitting parameters are shown in the top left corner for both months. All panels share the same color scale.

Comparison between C-GC and C-CC shows a different pattern. The line of best fit between C-CC and C-GC indicates 30% greater ozone in C-CC in January than in C-GC (y ~ 1.3x), but no such normalized mean bias is present in July (y ~ 1.0x). As with the comparison of C-GC to S-GC, the absolute bias is greater in July than in January, but the correlation between C-CC and C-GC does not worsen between the two months ($r^2 = 0.87$). This may indicate the strength of the effect of meteorology and non-chemistry processes in the seasonality of simulated surface ozone.

616 **4.3.   Aerosols**

617   Figure 6 shows the zonal mean mass concentration of sulfate aerosol as simulated in each of the three model configurations.

618   In C-GC and C-CC, this is calculated as the sum across all aerosol size bins, whereas S-GC uses a bulk representation.

619

620   *Figure 6. Comparison of sulfate aerosol mass concentration as simulated with CESM running GEOS-Chem (C-GC) to standalone GEOS-*
621   *Chem (S-GC) and CESM running CAM-chem (C-CC). Left (a): absolute values estimated with C-GC. Center (b): relative difference between*
622   *C-GC and S-GC. Right (c): relative difference between C-GC and C-CC. Red (blue) shading means that C-GC estimated a higher (lower)*
623   *value than the other model. Differences are restricted to ±100% for clarity.*

624   Between 45°S and 45°N, and below 800 hPa, C-GC more closely follows S-GC (comparison in panel b) with regards to sulfate

625   aerosol mass. Compared to C-CC (panel c), sulfate aerosol mass is approximately 50% greater in southern latitudes with

626   differences being greatest over the oceans. Sulfate concentrations in this region are dominated by oxidation of naturally-emitted

627   dimethyl sulfide (DMS) (Seinfeld and Pandis, 2006). Since DMS concentrations are identical between the three configurations,

628   the greater sulfate concentration in C-GC compared to C-CC may instead reflect differences in OH (Figure 3). Elsewhere the

629   concentration of sulfate in C-GC more closely follows that in C-CC, likely due to the common representation of sulfate aerosol

630   in MAM4 and differences in the representation of convective scavenging between CESM and standalone GEOS-Chem.

631   Concentrations of sulfate in the tropical mid-to-upper troposphere and extratropical lower stratosphere in C-GC exceed those

632   in S-GC by over 100%, whereas comparison to C-CC show differences of ±25%.

633

634

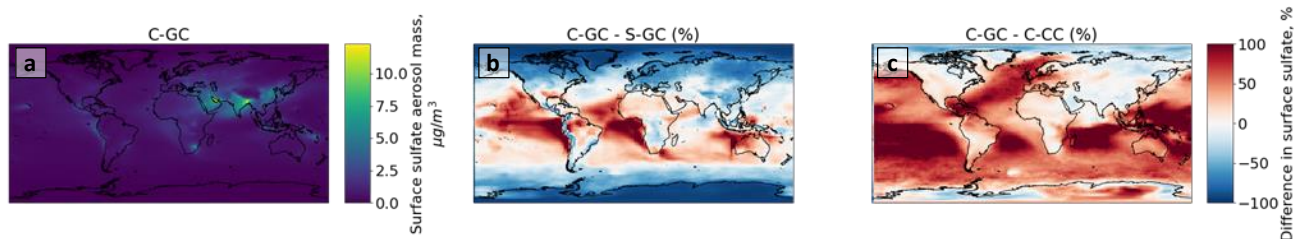



Figure 7. Comparison of the annually averaged surface mass concentration of sulfate aerosol simulated with CESM running GEOS-Chem (C-GC) to standalone GEOS-Chem (S-GC) and CESM running CAM-chem (C-CC). Red (blue) shading means that C-GC estimated a higher (lower) value than the other model.

638 This is further illustrated in Figure 7, which shows the surface concentration of sulfate aerosol in each model configuration.

639 C-GC simulated greater concentrations in the intertropical convergence zone (off the west coast of Southern Hemisphere

640 continents) than in S-GC (panel b), but in these regions agrees more closely with C-CC (panel c). Elsewhere in the tropics the

641 agreement between C-GC and S-GC is stronger, whereas surface concentrations of sulfate aerosol over (e.g.) the Southern

642 Pacific exceed those in C-CC by over 100%. At high latitudes and over land the agreement between C-GC and C-CC is again

643 stronger than in S-GC, although this varies by location. Further work would be needed to identify the underlying causes leading

644 to differences in surface sulfate concentrations between all three models.

645

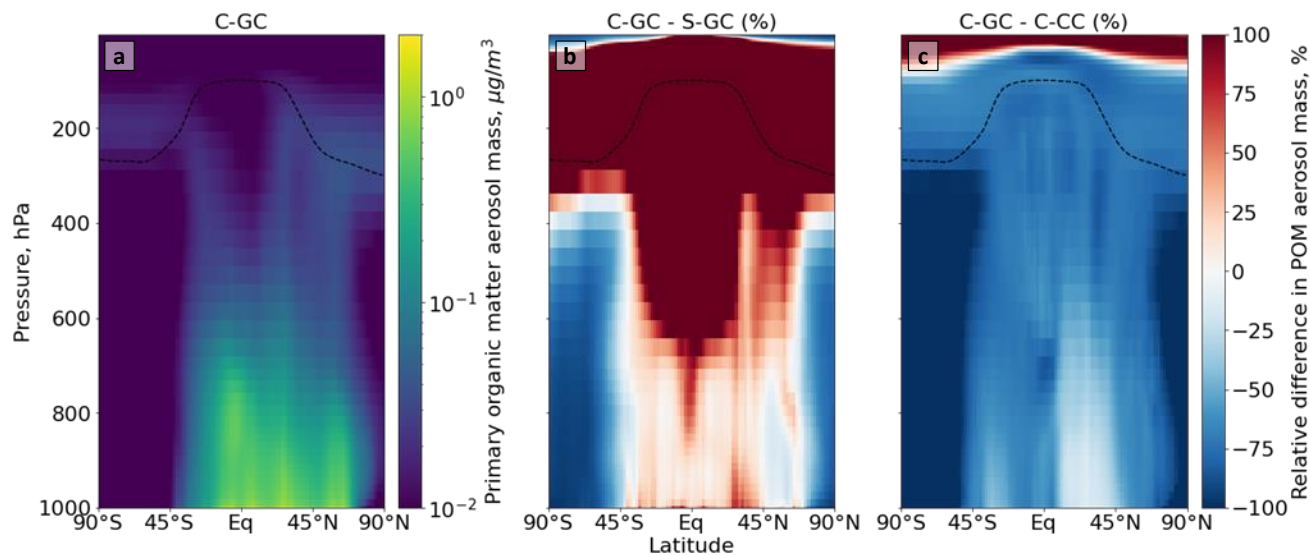



Figure 8. Comparison of primary organic matter aerosol mass concentration as simulated with CESM running GEOS-Chem (C-GC) to standalone GEOS-Chem (S-GC) and CESM running CAM-chem (C-CC). Left: absolute values estimated with C-GC. Center: relative difference between C-GC and S-GC. Right: relative difference between C-GC and C-CC. Red (blue) shading means that C-GC estimated a higher (lower) value than the other model.

650 We also show the zonal mean concentrations of primary organic matter (POM) aerosol in each configuration (Figure 8). POM

651 in C-GC and C-CC is calculated as the sum of the POM aerosol size bins, whereas in S-GC it is the sum of the hydrophobic

652 and hydrophilic organic carbon species. As with sulfate aerosol, C-GC and S-GC agree to within 25-50% in the tropics below

653   800 hPa, but C-GC simulates concentrations of POM which are over 100% greater than S-GC in the mid- and upper tropical

654   troposphere and throughout the lower stratosphere. This is again likely due to differences in the representation of convective

655   scavenging. C-GC also simulates concentrations of POM which are lower than C-CC throughout the entire troposphere. This

656   is likely due to differences in the implementation of POM emissions between C-CC and C-GC, where emissions of POM in

657   C-CC are 29% lower and occur as accumulation-mode rather than primary organic mode aerosol.

658   **4.4.   Reactive nitrogen (NO_y), bromine (Br_y), and chlorine (Cl_y)**

659   To better understand the source of differences in ozone and aerosols described above, we now investigate differences in

660   reactive nitrogen (NO_y) and halogen families (Br_y and Cl_y).

661   **4.4.1.   Reactive nitrogen (NO_y)**

662   We compare the total concentration and partitioning of reactive nitrogen species in each model configuration, including NO_x

663   and its reservoir species (collectively NO_y). A full list of the species included in the lumped NO_y reservoir species can be found

664   in the legend of Figure 10 for each model configuration. We first compare results in the stratosphere, followed by an evaluation

665   of concentrations and partitioning below 100 hPa. Concentrations of nitrate aerosol concentrations are estimated in CAM-

666   chem using a simplified approximation (Lamarque et al., 2012), and particulate nitrate is typically not considered to be

667   simulated by CAM-chem (e.g. Park et al. (2021)). We therefore do not include it in this analysis.

668

669

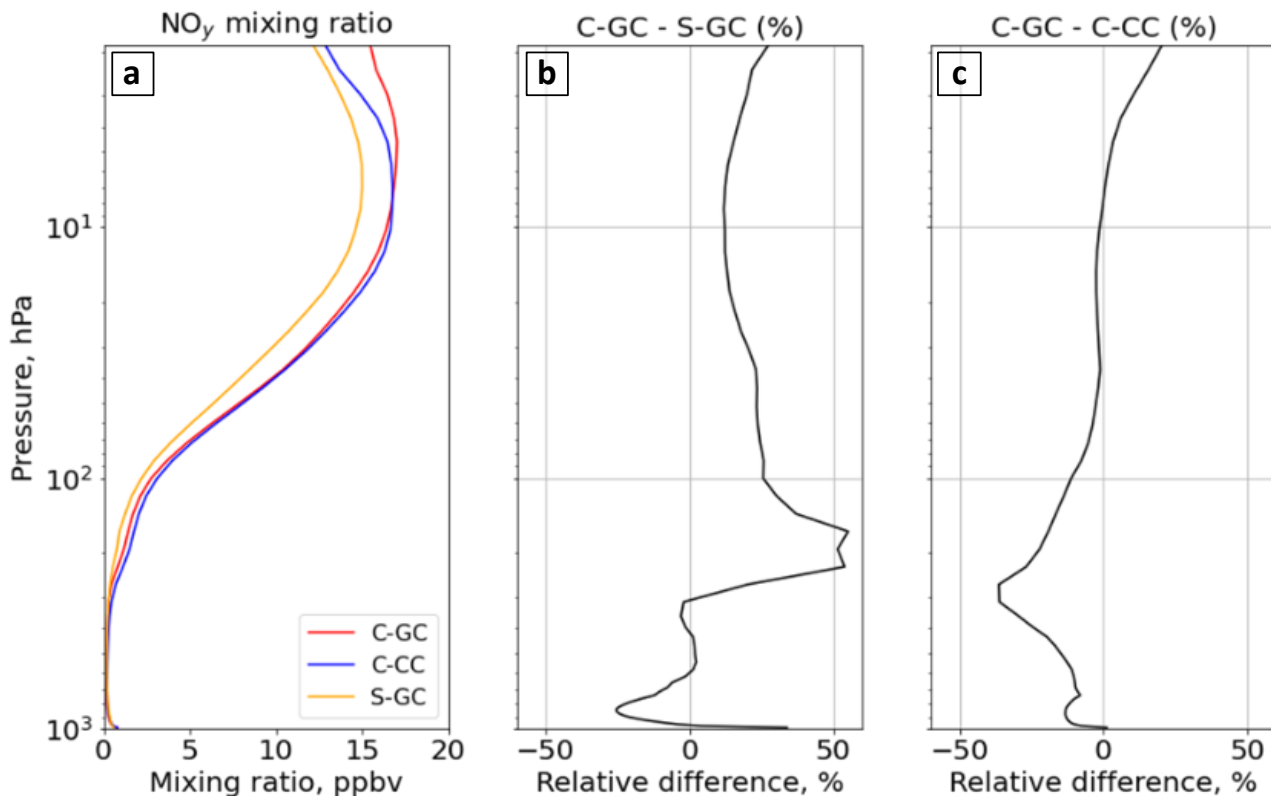



Figure 9. Global annual mean mixing ratio of total reactive nitrogen (NO_y) as a function of altitude. Left (panel a): Profile of NO_y mixing ratio for C-GC (red), C-CC (blue), and S-GC (orange). Middle (panel b): Relative difference in NO_y mixing ratio between C-GC and S-GC. Right (panel c): Relative difference in NO_y mixing ratio between C-GC and C-CC.

Figure 9 shows global mean NO_y at each altitude for C-GC, C-CC, and S-GC. Comparing C-GC to S-GC (panel b), differences in total NO_y are less than ±50% at all altitudes. Between 100 and 10 hPa, C-GC differs from S-GC by less than 20%, compared to less than 10% with respect to C-CC (panel c). The difference between C-GC and C-CC increases from -2% at 10 hPa to +20% at the top of the model, compared to an increase from 10% to 25% when comparing C-GC to S-GC. At lower altitudes C-GC more closely follows C-CC than S-GC, with differences between C-GC and S-GC exceeding 50% between 200 and 300 hPa. The global NO_y burden in C-GC (2.74 TgN) is closer to that in S-GC (2.84 TgN) than C-CC (3.01 Tg), likely due to the stronger influence of the troposphere on this quantity.

Figure 10 shows the speciation of NO_y as a function of altitude in each model from the surface to 1 hPa. The list of species defined collectively as NO_y differs between C-GC and S-GC on one side and C-CC on the other side. At altitudes above 100 hPa, the dominant contributors to NO_y in all three model configurations are NO, NO_2, HNO_3, and N_2O_5, although $ClNO_3$ contributes significantly between approximately 80 and 5 hPa. Between 10 and 200 hPa ratios of NO to NO_2 are approximately consistent between the models, lying in the range 0.35 to 0.50. This suggests broad consistency in actinic flux and ozone concentrations, given their role in controlling NO:NO_2 ratios in the stratosphere (Cohen and Murphy, 2003).

688   By contrast, partitioning between NO_x and HNO_3 differs significantly between the three models. At 10 hPa, HNO_3

689   constitutes 20% of total NO_y in C-GC but 23% in both C-CC and S-GC (values not shown explicitly). This fraction increases

690   with decreasing altitude at differing rates. At 200 hPa, HNO_3 constitutes 60 and 63% of NO_y in C-GC and S-GC

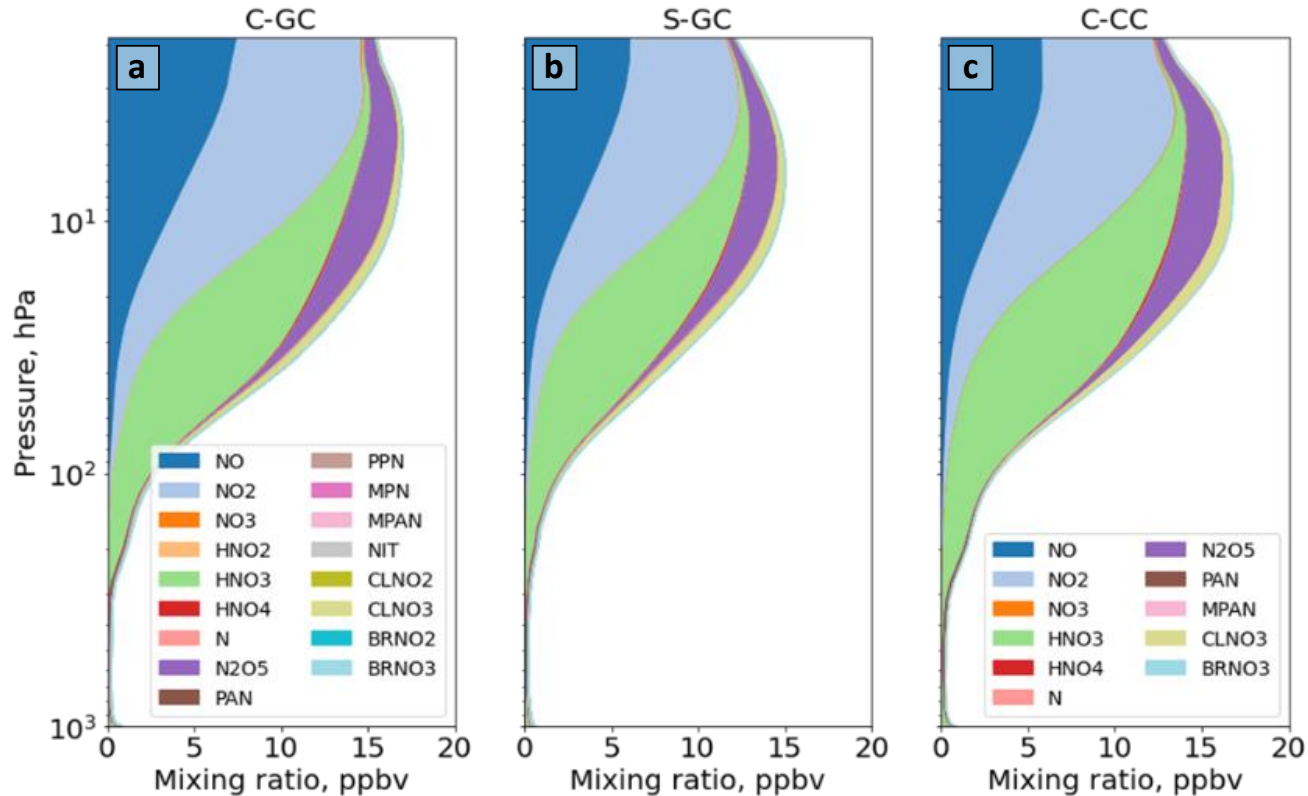
691   respectively, but 78% of NO_y in C-CC. One possible cause of these discrepancies is heterogeneous chemistry. GEOS-Chem

692   (in both S-GC and C-GC) uses a different representation of N_2O_5 hydrolysis than CAM-chem, but the CESM-driven

693   simulation includes a more detailed representation of the sulfate aerosol size distribution through MAM4. The present study

694   did not include the analysis of aerosol reactive tendencies. This would be a valuable line of inquiry for future comparisons of

695   CAM-chem and GEOS-Chem, given the lack of nitrate aerosol in the former.

696

697 *Figure 10. Global annual mean speciation of NO_y as a function of altitude. Results are shown from C-GC (left, a), S-GC (middle, b), and*
698 *C-CC (right, c) from the surface up to the model top (approximately 2 hPa). Values correspond to the number of N atoms present, such*
699 *that (e.g.) the mixing ratio of N_2O_5 is multiplied by 2.*

700   Figure 11 provides a closer look at the speciation of NO_y at altitudes below 100 hPa. NO_y at altitudes below 200 hPa is

701   predominantly NO_x, HNO_3, and peroxyacetyl nitrate (PAN). At 200 hPa, the combination of NO_x, HNO_3, and PAN make up

702   86% of total NO_y in C-GC (panel a) and 84% in S-GC (panel b), but 96% in C-CC (panel c). However, between 200 and 900

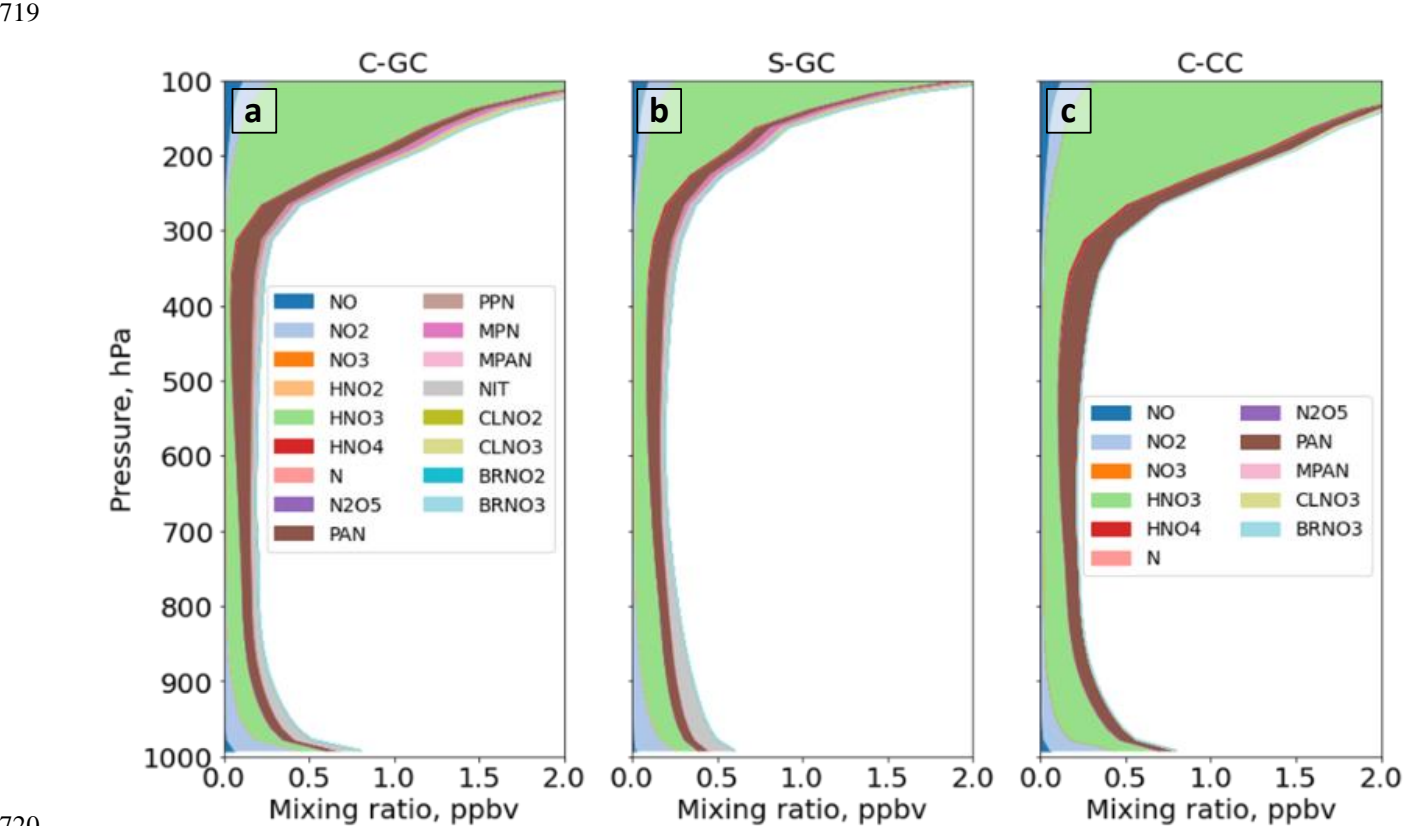
703   hPa the dominant contributors are HNO_3 and PAN. In this pressure range, the C-GC and S-GC simulations also show a

704   significant contribution from nitrate aerosol (NIT) and $BrNO_3$. At 500 hPa, the contributions of NO_x, HNO_3, and PAN are

705 78%, 85%, and 97% respectively for C-GC, S-GC, and C-CC. Below 900 hPa, NO and NO_2 once again become significant
706 contributors to total NO_y. At these lower altitudes C-GC more closely follows C-CC than S-GC, with differences in total NO_y
707 between C-GC and S-GC exceeding 50% between 200 and 300 hPa. As such, we find that the speciation in C-GC more closely
708 follows that in S-GC at lower altitudes.

709

710 Since surface emissions of NO_x are nearly identical between the three configurations (see Table 2) and lightning NO_x emissions
711 are identical between C-GC and C-CC, differences below 100 hPa may instead be related to NO_x chemistry, and nitrate aerosol.
712 However, concentrations of PAN in C-GC more closely follow C-CC than S-GC, suggesting that the representation of
713 meteorology (including wet deposition rates) is also an important factor. At 500 hPa, total PAN in C-GC is 3% lower than the
714 value in C-CC, but exceeds the value in S-GC by 38%. This may be due to the greater emissions of biogenic VOCs in CESM
715 than in the standalone GEOS-Chem (see Table 3Table 3), resulting in more NO_x being bound into PAN for long-range
716 transport. We also find that HNO_3 concentrations in the mid-troposphere are lower in C-GC than in either C-CC or S-GC. At
717 500 hPa, HNO_3 mixing ratios in C-GC are 43% lower than in S-GC and 52% lower than in C-CC. This does not account for
718 the conversion of HNO_3 in C-GC and S-GC to nitrate aerosol (NIT), which is not represented in C-CC.

719

720
721 *Figure 11. As in Figure 10, but showing only the 10^3-10^2 hPa pressure range.*

722 Differences in mid-tropospheric HNO_3 between the models are most likely due to differences in the representation of wet
723 scavenging. In C-CC and C-GC, scavenging of gaseous species is handled by the Neu scheme, while scavenging of modal
724 aerosols is performed by MAM (Neu and Prather, 2012). Any aerosol species not handled by MAM, such as nitrate in C-GC,
725 are also scavenged using the Neu scheme. In C-GC and C-CC, the Neu scheme calculations are performed at the same time as
726 the chemistry and after convective transport, while scavenging of MAM aerosols is performed before. Thus, all species that
727 undergo wet deposition in the Neu scheme are not removed during convective transport. This leads to soluble species and
728 aerosols being carried to higher altitudes without being convectively scavenged. We also find that the Neu scavenging scheme
729 in C-GC and C-CC results in an HNO_3 wet removal rate which is four times higher in C-GC than in S-GC (Figure S1 of the
730 Supplementary Information). This likely explains the greater depletion of HNO_3 in the mid-troposphere calculated by C-GC
731 compared to S-GC. Wet scavenging in C-CC is faster yet, with HNO_3 wet removal rates approximately six times greater than
732 in S-GC, and 50% greater than in C-GC. This is in part because the mixing ratio (or fraction of total NO_y) of HNO_3 in the mid-
733 and upper-troposphere as modeled in C-CC is greater than in either C-GC or S-GC, but also because C-GC and S-GC simulate
734 nitrate aerosol explicitly. The application of the Neu scheme to remove nitrate aerosol also affects removal of total NO_y in C-
735 GC (Figure S2 in the Supplementary Information). We find that the Neu scheme removes aerosol more rapidly than the
736 scheme used in S-GC, and at lower altitudes.
737
738 Total HNO_3 removal tendencies in each model configuration are shown in Table 7. The total removal rate of NO_3^- is lowest in
739 S-GC and highest in C-CC, consistent with the finding that total NO_y burdens are lower in S-GC than C-GC or C-CC. However,
740 the removal rate of nitrate aerosol is lower in C-GC than in S-GC despite the greater wet removal rates for C-GC. A possible
741 explanation is that washout rates of nitrate aerosol are sufficiently high in both C-GC and S-GC that all nitrate aerosol is
742 effectively removed, but that the faster washout of HNO_3 in C-GC results in less nitrate aerosol being available for removal.
743
744 *Table 7. Total wet removal tendency of HNO_3 and nitrate aerosol in each model configuration. All values are given in units of Tg NO_3/yr.*

	C-GC	S-GC	C-CC
HNO_3	82.0	71.3	119.6
Nitrate aerosol	20.4	22.7	-
Total NO_3^-	102.4	94.0	119.6

745

746 **4.4.2.   Reactive bromine (Br_y)**

747 Halogens are involved in multiple catalytic ozone-depleting chemical cycles, and are critical to an accurate description of both
748 tropospheric and stratospheric chemistry (Solomon et al., 2015). We therefore analyze the abundance and speciation of the
749 two key halogens – bromine and chlorine – in each configuration. Figure 12 shows the annual average mixing ratio of total
750 reactive bromine as a function of altitude in each of the three models. This does not include long-lived species such as halons
751 or CH_3Br. A full listing is included in the legend of Figure 13.

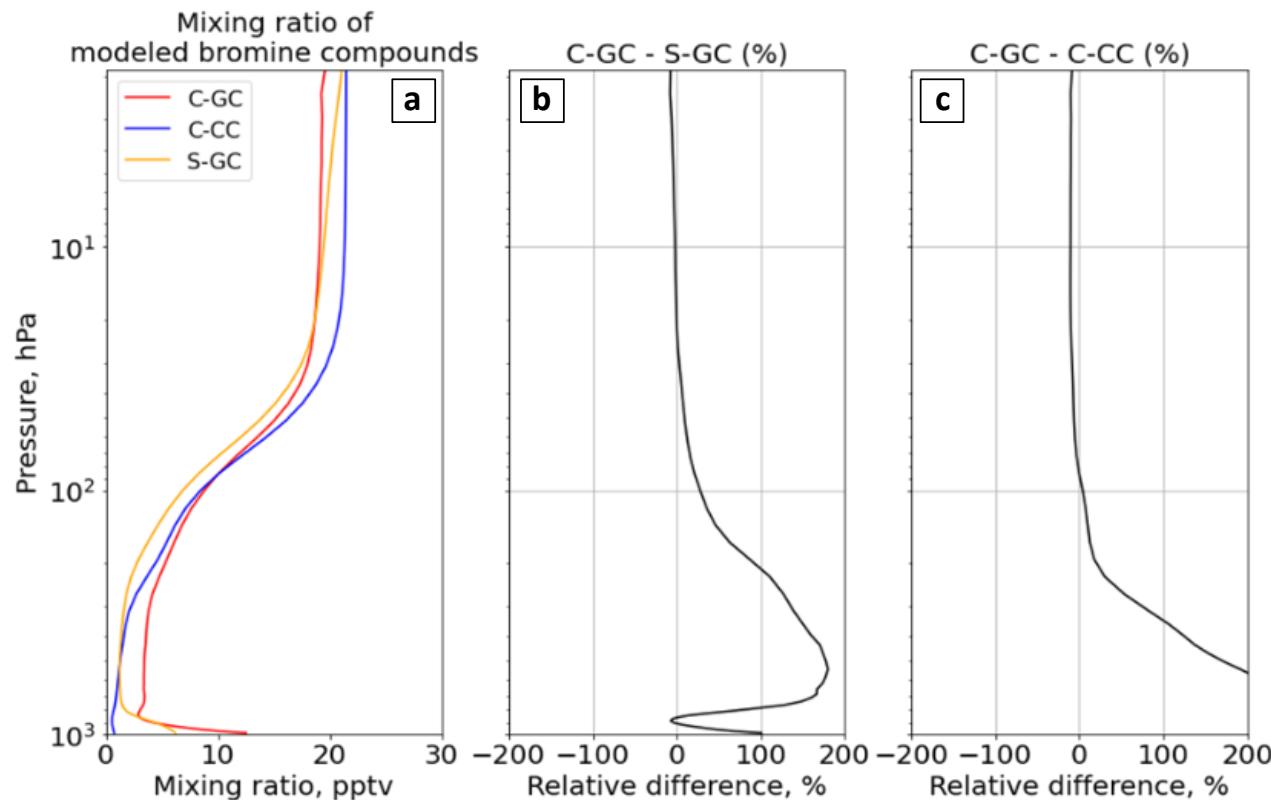



Figure 12. Global annual mean mixing ratio of reactive bromine as a function of altitude. Left (panel a): Profile of total gaseous inorganic and organic bromine mixing ratio for C-GC (red), C-CC (blue), and S-GC (orange). Middle (panel b): Relative difference in bromine-containing species mixing ratio between C-GC and S-GC. Right (panel c): Relative difference in bromine-containing species mixing ratio between C-GC and C-CC. Although relative differences between C-GC and C-CC exceed 1000% near the surface, the limits on the rightmost panel are clipped to allow comparison to the center panel.

Globally averaged total Br_y in C-GC is maximized at the surface, exceeding that from S-GC by 100%. This is partially explained by the greater emissions of sea salt bromine, although C-GC's annual emission of sea salt bromine is only 36% greater than that in S-GC (see Table 4). Since C-CC does not include short-lived bromine sources such as sea salt bromine, the C-GC total Br_y concentration exceeds C-CC by 1000 % at the surface.

From Figure 13, we find that, in all three models, the mixing ratio increases monotonically with altitude above 800 hPa due to the reaction of CH_3Br with OH. Br_y falls sharply from 12 pptv at the surface in C-GC to 3 pptv at 900 hPa, but then increases again to 10 pptv at 100 hPa. This pattern is similar to that displayed by S-GC, although the decrease from the surface is less sharp and the absolute value lower in S-GC. Above 100 hPa, the averaged Br_y mixing ratio levels off, with values between 20 hPa and 2 hPa remaining roughly constant in the range of 16-20 pptv. This is similar to the behavior shown by C-CC but differs from S-GC, in which Br_y continues to rise with altitude – albeit more slowly. The net effect is that total Br_y in C-GC

770 exceeds both C-CC and S-GC below 100 hPa, but is lower than the value in either model above 10 hPa (above 80 hPa when

771 compared to C-CC).

772

773 In addition to differences in total Br_y, the partitioning of Br_y also varies between the three models (Figure 13). The additional

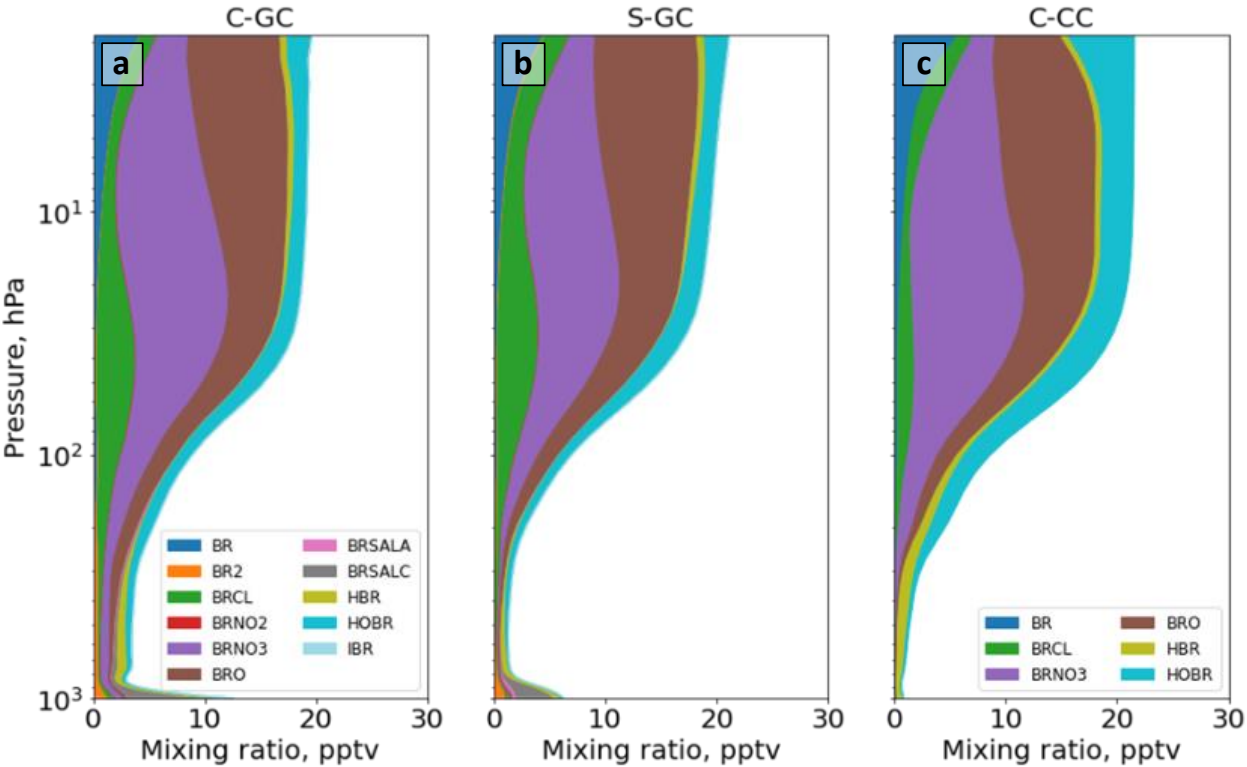
774 near-surface bromine present in C-GC and S-GC is due to the presence of Br_2 and sea salt bromine (BrSALA and BrSALC,

775 representing bromine in fine and coarse-mode sea salt respectively). This provides a source of active bromine in the planetary

776 boundary layer which is not represented in C-CC, but in forms which are rapidly washed out in C-GC and S-GC. The greater

777 concentrations of Br_y near the surface as calculated by C-GC compared to S-GC are likely due to the greater emissions of sea

778 salt bromine, as shown in Table 4.

779

780

781 *Figure 13. Global annual mean speciation of total organic and inorganic bromine as a function of altitude. Results are shown from C-GC*
782 *(left, a), S-GC (middle, b), and C-CC (right, c), from the surface up to the model top (approximately 2 hPa). Values correspond to the*
783 *number of Br atoms present, such that (e.g.) the mixing ratio of Br_2 is multiplied by 2.*

784 Br_y in the model stratosphere is dominated by the same species in all three configurations: BrO_x (Br + BrO), BrCl, $BrNO_3$,

785 HBr, and HOBr. The most significant difference is the greater proportion of HOBr in C-CC (approximately 15%) than in S-

786 GC or C-GC (8-10%). Larger mixing ratios of $BrNO_3$ are also present in C-CC (approximately 10 ppbv at 30 hPa) compared

787 to C-GC and S-GC (approximately 7.4 and 7.0 ppbv respectively at 30 hPa). Smaller mixing ratios of BrCl are present in C-

788 CC, with a mean value of 1.8 ppbv at 30 hPa, while they reach 3.1 ppbv in C-GC and S-GC at 30 hPa. The base causes of

789 these differences are not clear, but may be related to the presence of more complex tropospheric and stratospheric halogen
790 chemistry in the GEOS-Chem chemical mechanism (Wang et al., 2021).

792 Between 30 hPa and the top of the boundary layer, the three models show greater divergence. The only significant sources of
793 atmospheric Br_y in C-CC are CH_3Br, CH_2Br_2, and very long-lived bromine species such as halons which are insoluble. As a
794 result, tropospheric Br_y concentrations increase only slowly from the surface up to 300 hPa, at which point HOBr, BrO, and
795 $BrNO_3$ begin to form in significant quantities. In C-GC and S-GC, these sources of bromine are supplemented by bromine
796 from sea salt and surface Br_2 emissions. Mid-tropospheric Br_y concentrations are therefore largely set by the quantity of sea
797 salt bromine emitted, and by the fraction of that bromine which can be released to an insoluble form (e.g. Br_2) before the sea
798 salt is washed out of the atmosphere.

800 The greater concentration of mid-tropospheric Br_y in C-GC than in S-GC is likely due to differences in wet scavenging. Wet
801 removal tendencies of bromine in fine sea salt (BrSALA) from large-scale and convective precipitation as calculated by C-GC
802 and S-GC are shown in the Supplementary Information (Figure S3). We find that there is greater wet deposition of fine sea
803 salt bromine in S-GC than in C-GC, despite removal rates below 900 hPa being greater in C-GC. Since total emissions of
804 BrSALA are also 26% lower in S-GC than in C-GC (Table 4), the slower mid-tropospheric mid-tropospheric removal of
805 bromine in C-GC explains the greater simulated concentration of Br_y in the mid troposphere.

807 C-GC also calculates wet deposition of non-MAM aerosols from both convective and large-scale precipitation independent of
808 convective transport, whereas S-GC calculates convective scavenging as part of convective transport. This means that soluble
809 species can be transported in convective updrafts in C-GC, unlike in S-GC.

4.4.3. Reactive chlorine (Cl_y)

811 We now focus on atmospheric chlorine by comparing its profile and partitioning in all three models. Annually-averaged
812 vertical profiles of reactive chlorine (Cl_y) are displayed in Figure 14, excluding source species such as chlorocarbons. A full
813 list of the species used to define Cl_y in each configuration is provided in Figure 15.

815 As with total Br_y, total Cl_y follows the same vertical distribution as S-GC up to 10 hPa. Above this pressure, the vertical
816 distribution in C-GC is closer to that of C-CC. The dominant factor in differences below 100 hPa is the lack of short-lived
817 chlorine species such as sea salt in C-CC, which are the dominant source of chlorine to the lower troposphere. Above 10 hPa,
818 the relative difference in Cl_y between C-GC and S-GC increases slowly from -2% at 10 hPa to -5% at 2 hPa, while the difference
819 relative to C-CC remains at approximately -20% above 200 hPa.

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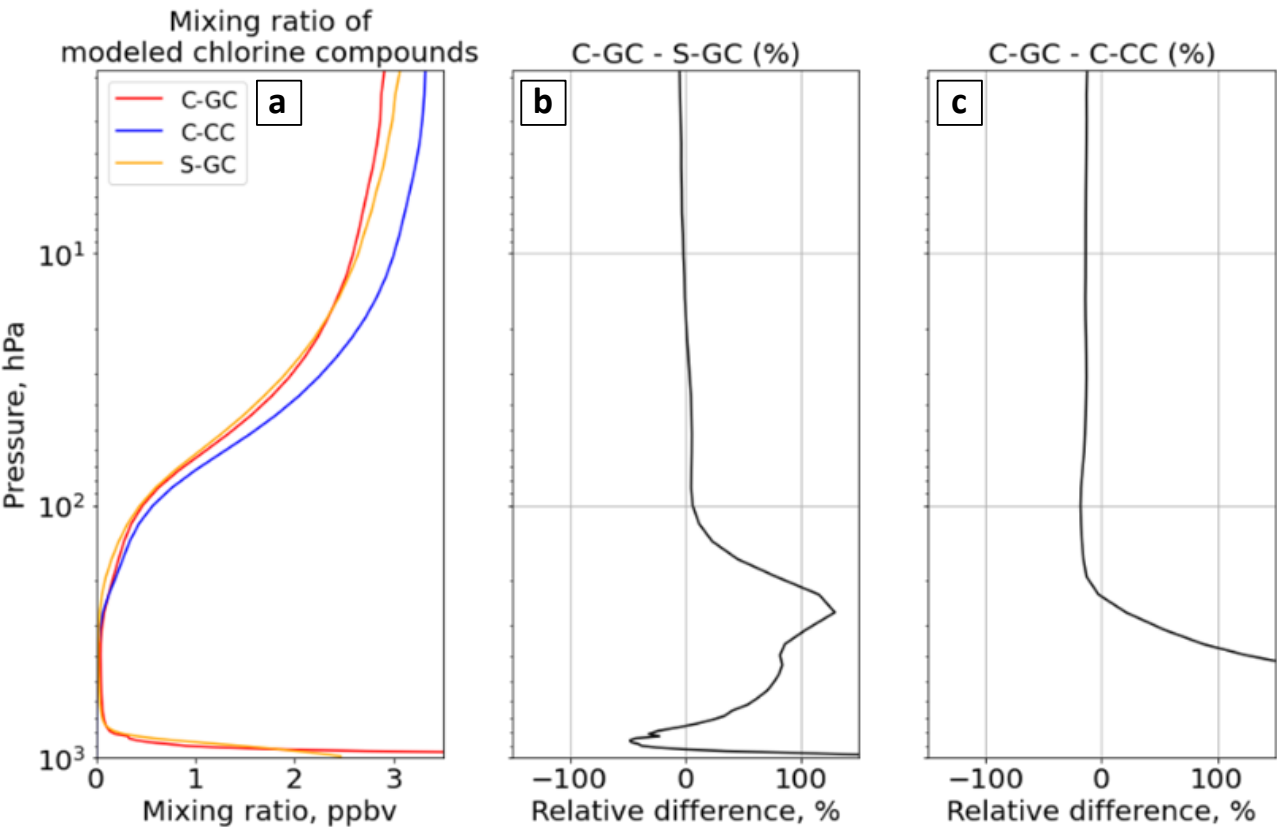
821

822 *Figure 14. Comparison of annual average vertical profiles of chlorine-containing compounds in the three models. Left (panel a): Vertical*
823 *profile of total gaseous chlorine mixing ratio for C-GC (red), S-GC (orange), and C-CC (blue). Middle (panel b): Relative difference in*
824 *Cl_y mixing ratio between C-GC and S-GC. Right (panel c): Relative difference in Cl_y mixing ratio between C-GC and C-CC. Although*
825 *relative differences between C-GC and C-CC exceed 1000% near the surface, the limits on the rightmost panel are clipped to allow*
826 *comparison to the center panel.*

827 Figure 15 shows the speciation of Cl_y as a function of altitude in each model. The greater near-surface chlorine simulated by

828 C-GC (panel a) and S-GC (panel b) relative to C-CC (panel c) is mostly made up of HCl and chlorine in sea salt (SALACL

829 and SALCCL). However, the short lifetime of sea salt aerosol and of chlorine in the lower troposphere means that this source

830 is likely not significantly affecting stratospheric chlorine burdens or partitioning. In the stratosphere there is no clear difference

831 between partitioning in C-GC and S-GC, but larger mixing ratios of upper tropospheric and lower stratospheric HCl from

832 chlorine source compounds are found in C-CC. At 50 hPa, total Cl_y in C-CC is 15% greater than in C-GC and S-GC, but the

833 mean mixing ratio of HCl in C-CC is 45% greater. Differences in $ClNO_3$ reach their peak at higher altitudes, around 20-30

834 hPa.
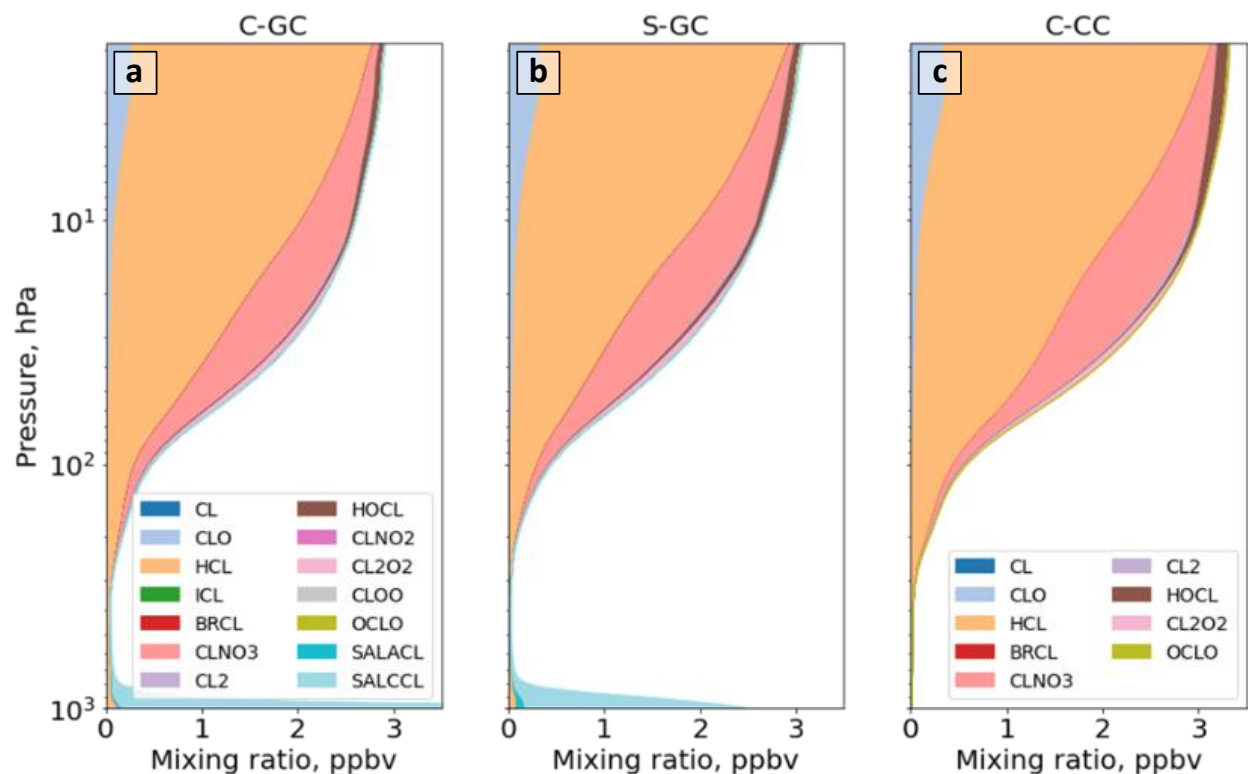
Figure 15. Global annual mean vertical speciation of total organic and inorganic bromine in C-GC (left, a), S-GC (middle, b), and C-CC (right, c) from the surface up to the model top (approximately 2 hPa). Values correspond to the number of Cl atoms present, such that (e.g.) the mixing ratio of Cl_2 is multiplied by 2. SALACL and SALCCL correspond to chlorine in fine and coarse sea salt, respectively.

The global mean tropospheric concentration of Cl atoms is 590 cm^{-3}, roughly consistent with a recent evaluation from Wang et al. (2021) which found a value of 630 cm^{-3}. This is 24% greater than the value from S-GC (477 cm^{-3}) and 160% greater than that from C-CC (224 cm^{-3}), likely due to the greater emissions of sea salt and indicating that chlorine will play a greater role in tropospheric oxidation in C-GC.

5.   Comparison of model results to observations

We now compare the results from C-GC to observational data, with results from S-GC and C-CC also provided as context. Section 5.1 evaluates model performance at the surface, comparing to ground measurements of surface NO$_2$ and ozone. Section 5.2 compares model results to a climatology of vertical profiles of ozone, based on ozone sonde data. Section 5.3 evaluates the level of agreement of simulated ozone and carbon monoxide (CO) columns to measurements from the Aura Ozone Monitoring Instrument (OMI) and Microwave Limb Sounder (MLS), and MOPITT satellite instruments. Finally, Section 5.4 evaluates the model against measurements of dry deposition fluxes and rainwater composition measurements. The CESM wet deposition

scheme is presented in Neu and Prather (2012), while the wet deposition scheme implemented in GEOS-Chem uses different schemes for gases and aerosols (Liu et al., 2001; Amos et al., 2012). Dry deposition in GEOS-Chem is described in several publications (Wesely, 1989; Wang et al., 1998; Zhang et al., 2001), while the coupling between CAM and CLM is described in Val Martin et al. (2014) .

5.1. Surface NO$_2$ and ozone

Figure 16 compares surface mass concentrations of NO$_2$ as estimated by C-GC, S-GC, and C-CC for 2016 against ground station measurements for North America (top row), Europe (middle row), and South-East Asia (bottom row) (AirNow API, 2021; Environmental Numerical Database, 2021; China Air Quality Historical Data, 2021; European Air Quality Portal, 2021). All ground station measurements are the average value over 2016. By comparing model results at an approximately 2° horizontal resolution to point observations, we expect some differences in this evaluation due to grid-box representation errors.

All three model configurations calculate lower mixing ratios than are reported by the ground observations. This is likely to be in part due to the presence of interferants such as HNO$_3$, which cause in-situ monitors to overestimate the concentration of NO$_2$ (Dunlea et al., 2007). However, S-GC is consistently biased lower than C-GC or C-CC. We also find that the surface NO$_2$ concentrations display variable agreement depending on the geographical location. The correlation coefficients for North America, Europe and South-East Asia are provided in Table 8. All three models give similar correlation coefficients for each region. This is expected given that the three model configurations all use the same input wind fields and NO$_x$ emissions datasets. Nonetheless, both C-GC and C-CC estimate higher concentrations of NO$_2$ in Northern China, Northern Europe, and the northeast US than S-GC. This suggests that the representation of meteorology, photolysis, and NO$_y$ removal processes have a greater impact on simulated NO$_2$ than the chemistry module alone. Comparing in-situ NO$_2$ measurements against NO$_y$ model results could potentially remove the effect of interferants in the observations of surface NO$_2$ concentrations, but is not considered here.

Table 8. Correlation coefficients of surface-level NO$_2$ mass concentrations for C-GC, S-GC and C-CC against measurements.

	C-GC	S-GC	C-CC
North America	0.39	0.36	0.38
Europe	0.21	0.21	0.21
South-East Asia	0.42	0.41	0.41

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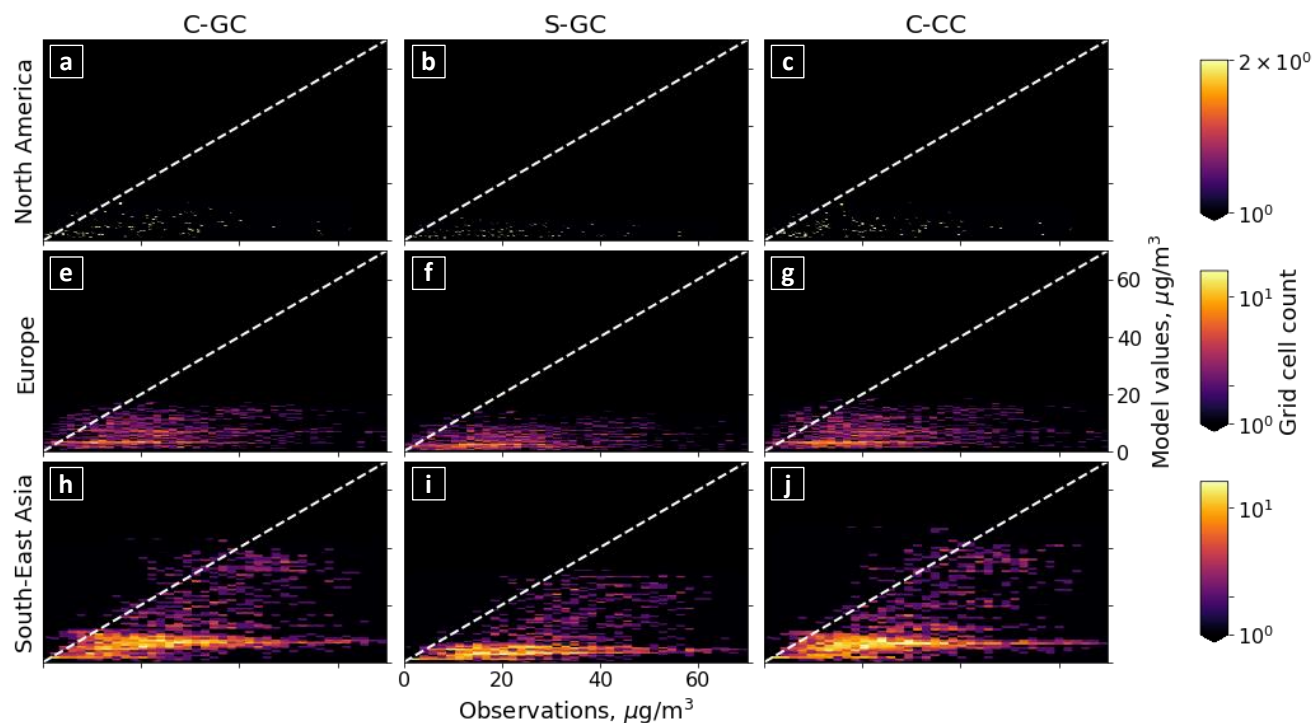
Figure 16. Annual average surface NO$_2$ mass concentrations simulated by C-GC (left: a, e, h), S-GC (middle: b, f, i), and C-CC (right: c, g, j) for 2016 compared against monitor measurements in North America (top: a-c), Europe (middle: d-e), and South East Asia (bottom: h-j).

Figure 17 shows the ratio of annual mean NO to annual mean NO$_2$, and thus provides some insight into possible causes of these disagreements. The global distribution of surface-level NO2 and NOx is presented in the Supplementary Information. All three configurations show enhanced NO:NO$_2$ ratios in polluted regions such as eastern China and over icy regions such as Greenland and Antarctica. However, S-GC shows reduced NO:NO$_2$ ratios over land compared to either C-CC or C-GC. For example, ratios over North America in S-GC range from 0.1 to 0.2, compared to a range of 0.01 to 0.1 in C-GC and C-CC. Surface NO:NO$_2$ ratios are typically dictated by surface ozone and the NO$_2$ photolysis rate (Seinfeld and Pandis, 2006). Given that surface ozone concentrations in S-GC are typically between those calculated in C-GC and C-CC (see Figure 4) and that S-GC and C-GC share the same photolysis treatment, this discrepancy may instead be caused by the differences in cloudiness calculated by CESM compared to the MERRA-2 fields read in by S-GC.
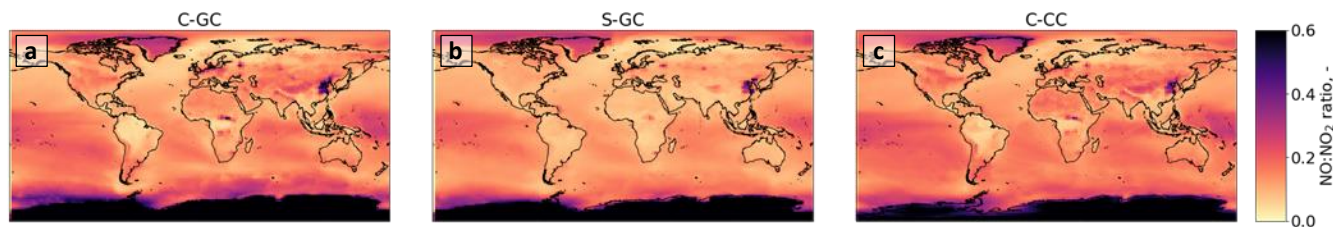
Figure 17. Surface-level NO:NO₂ estimated by C-GC (left, a), S-GC (middle, b), and C-CC (right, c) for 2016. Annual average of NO:NO₂ is calculated as annual mean NO divided by annual mean NO₂.

Differences in NO:NO₂ may also be related to differences in emissions and treatment of oxidants such as VOCs and bromine. C-GC and C-CC show a reduction in NO:NO₂ over the Amazon and in the Congo river basin, but this pattern is not reproduced in S-GC. Similarly, topographical features including the Andes and Himalayas are visible in the C-CC and C-GC NO:NO₂ ratios, but not in the S-GC data – whereas a large reduction in NO:NO₂ over the Arctic Ocean is more pronounced in S-GC and C-GC than in C-CC. This latter feature may be related to differences in the response of the simulated atmosphere to anthropogenic emissions, as ship tracks are more visible in the C-GC and S-GC NO:NO₂ ratios (see e.g. Cape Horn and the Cape of Good Hope) than in C-CC.

Figure 18 compares surface ozone against monitor measurements. The correlation coefficients are shown in Table 9. The geographical pattern is also consistent, with high surface ozone concentrations over the Mediterranean Sea and lower concentrations over Northern Europe.

Table 9. Correlation coefficients of surface-level ozone mass concentrations for C-GC, S-GC and C-CC against measurements.

	C-GC	S-GC	C-CC
North America	0.37	0.28	0.24
Europe	0.44	0.43	0.44
South-East Asia	-0.24	-0.07	-0.22

However, the results from all models appear to be biased low. As discussed in Section 4.2, C-GC estimates surface ozone mixing ratios lower than either S-GC and C-CC, and therefore exhibits the greatest mean bias. C-GC, C-CC, and S-GC show mean biases of -15, -9, and -10 ppbv for over Europe; -10, -3, and -5 ppbv over North America; and -20, -11, and -12 ppbv over South-East Asia.
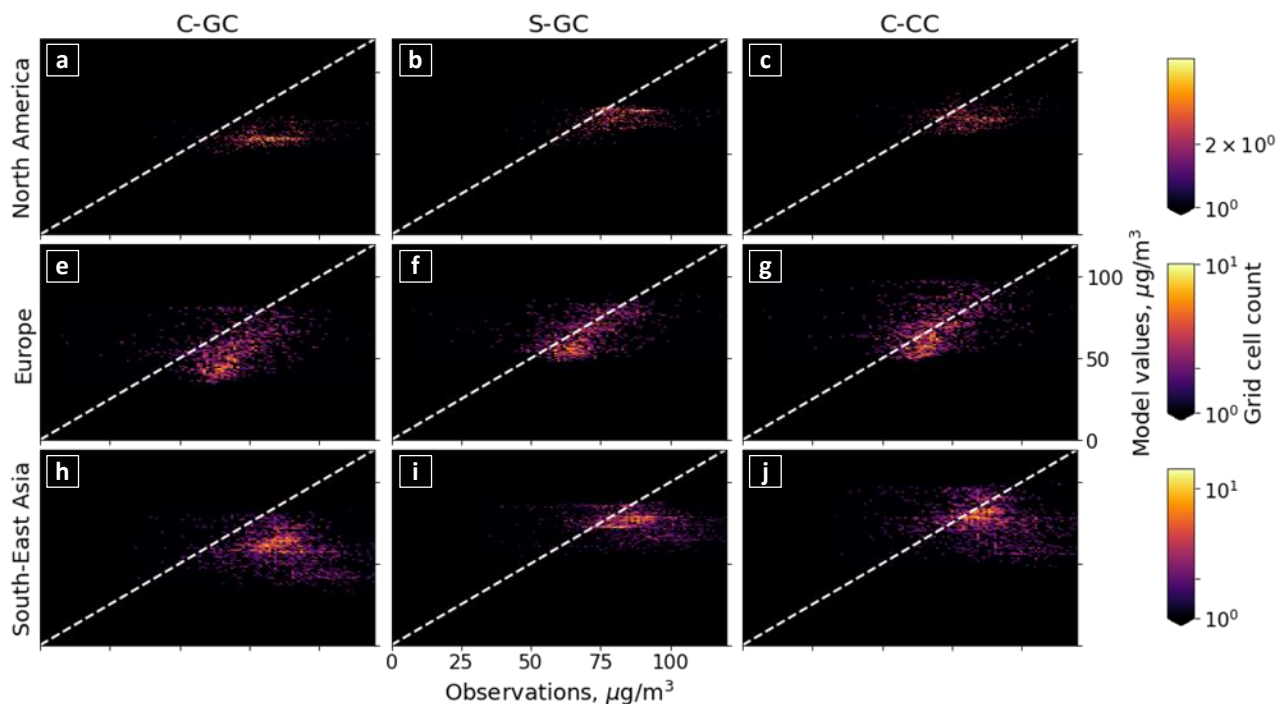
Figure 18. Annual average surface ozone mass concentrations simulated by C-GC (left: a, e, h), S-GC (middle: b, f, i), and C-CC (right: c, g, j) for 2016 compared against monitor measurements in North America (top: a-c), Europe (middle: d-e), and South East Asia (bottom: h-j).

The greater negative bias in simulated ozone shown by C-GC is likely related to both the different representation of meteorology compared to S-GC and the greater bromine emissions compared to both S-GC and C-CC. However, further work is needed to disentangle the root causes of discrepancies between the three models, and the common biases relative to observations.

5.2. Vertical profiles of ozone

We now focus on the evaluation of the profile of ozone mixing ratios by comparing C-GC, C-CC, and S-GC to a climatology of ozone sonde observations from 1995-2010 (Tilmes et al., 2012). This climatology (in addition to those shown in Section 5.3) has been used repeatedly in evaluations of CAM-chem, and is therefore chosen here to provide a familiar point of comparison which also allows the three configurations to be compared to a realistic baseline. Over the past decades, observations from ozone sondes in different locations provide a valuable dataset of the evolution of ozone mixing ratios in the troposphere and stratosphere. Figure 19 provides a Taylor diagram comparison between the C-GC, C-CC, and S-GC simulations of 2016 to the climatology.

929   In general, C-GC does not perform significantly better or worse than C-CC or S-GC, producing mean biases and correlations

930   in each region/altitude combination which are within the same range. The clearest exception is at low altitudes (900 or 500

931   hPa) and mid- to high latitudes. In these regions, C-GC results frequently show a smaller normalized difference from the mean

932   (radius) than either S-GC or C-CC, but also a weaker correlation with the observed seasonal cycle. The C-GC simulation of

933   tropical ozone also shows the smallest mean bias at all altitudes at or below 250 hPa (panels a, d, and g), although again

934   showing a weaker correlation.

935

936   At high altitude (50 hPa, panels j-l), all three models appear to perform similarly. This may however simply reflect the lack of

937   spin up time. Since the three models only simulated 1.5-2 years in total, the simulated stratosphere will not have had time to

938   fully respond to the new model configuration. Longer simulations are needed to fully evaluate the performance and capability

939   of the C-GC stratosphere.

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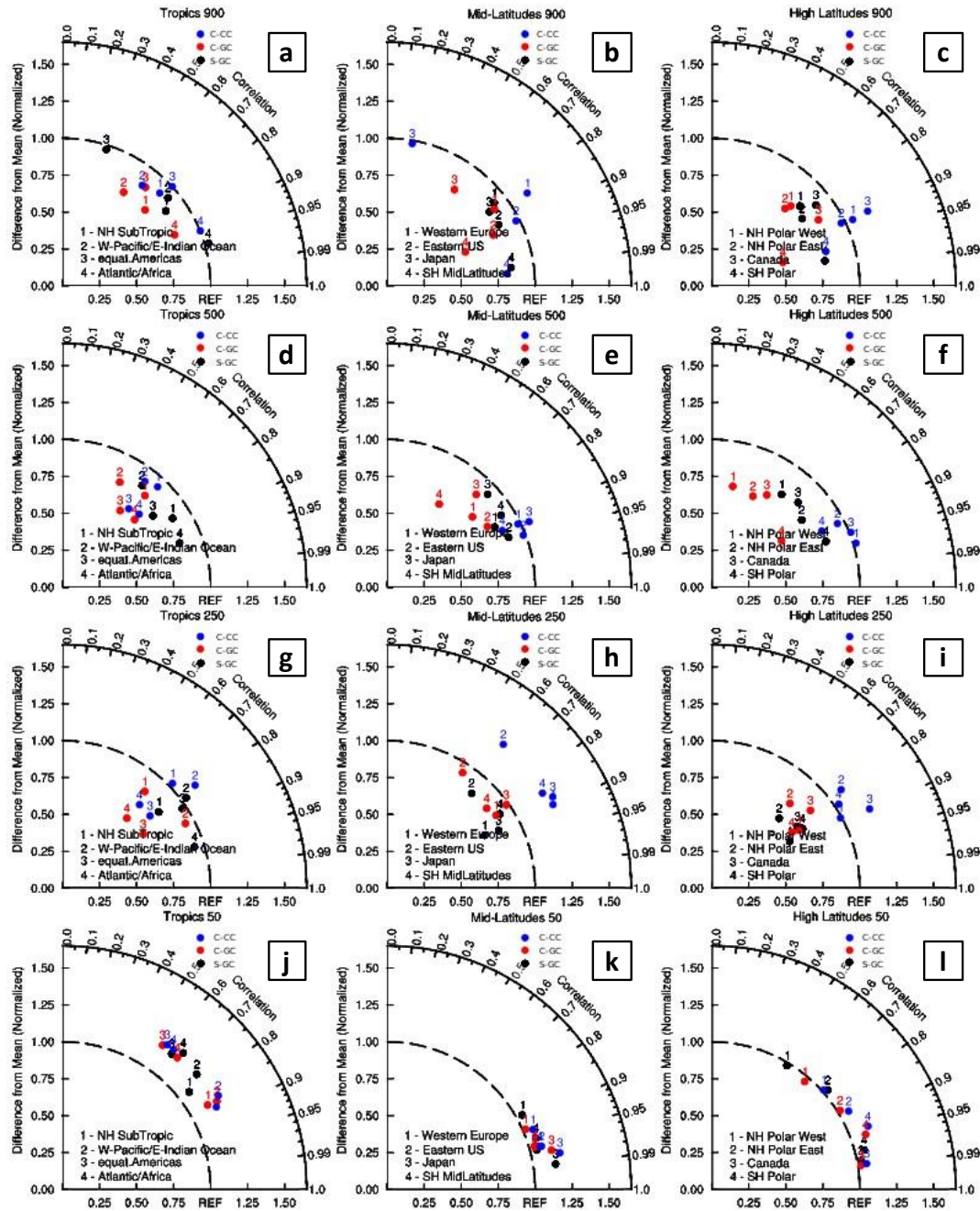
Figure 19. Taylor diagrams of the comparison of C-GC (red), C-CC (blue), and S-GC (black) simulations to a present-day (1995-2010) ozone sonde climatology. Top row to bottom row: comparisons at 900 (a-c), 500 (d-f), 250 (g-i) and 50 hPa (j-l). Left column to right column: tropics (a, d, g, j), mid latitudes (b, e, h, j), and high latitudes (c, f, i, l). The normalized mean difference between simulations and observations for each region is shown on the radius, and the correlation of the seasonal cycle is shown as the angle from the vertical.

5.3. Total column ozone and CO

Figure 20 shows total ozone column climatologies in Dobson Units as measured by OMI/MLS for the 2004-2010 time period (Ziemke et al., 2011). The results from the satellite observations (panel a) are compared to results from C-GC, C-CC, and S-GC (panels b, d, and f respectively,). We find that on average the results from C-GC are 7.8 DU lower than the observations (panel c), mostly driven by an overestimation of stratospheric ozone depletion during the Antarctic spring of up to 16 DU (value not shown in figure). C-CC predicts a total ozone column that is 6.6 DU larger than the global mean ozone column (panel e). When broken down by tropospheric and stratospheric ozone column, we find that the bias in the stratospheric and tropospheric ozone columns for C-GC is -2 and -6 DU respectively, compared to +9.5 and -2.5 DU for C-CC (value not shown in figure). Additionally, we find that the bias in seasonal variations of total column ozone as predicted by C-GC range between -16 and -6 DU, while the variations range from -3 to +7 DU for C-CC (value not shown in figure). The model results from S-GC predict similar geographical biases in total ozone column as C-GC, although with a smaller net bias of -3.3 DU (panel g).

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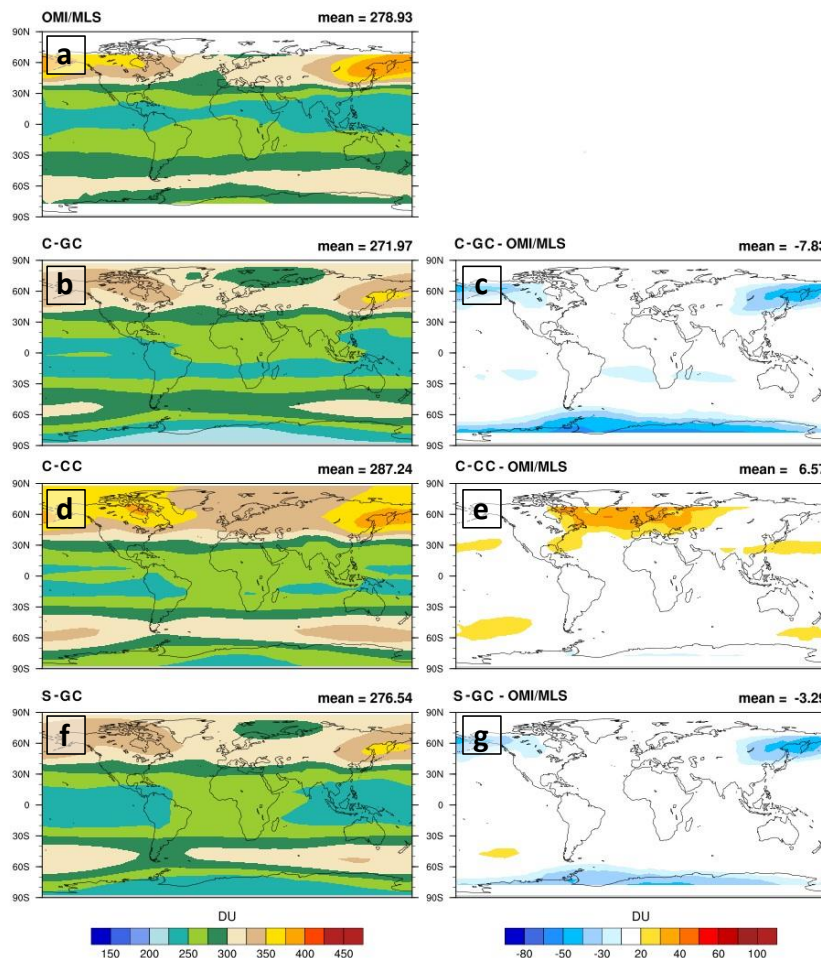
Figure 20. Total ozone column in DU as observed by OMI/MLS for the 2004-2010 time period (panel a), compared to the results from C-
GC (2nd row, panels b and c), C-CC (3rd row, panels d and e), and S-GC (4th row, panels f and g) for the year 2016. The measurements
and model results are presented on the left (a, b, d, f), while the model biases are shown on the right (c, e, g).

961

Figure 21 compares the simulated total columns of CO (panels b, d, and f) to retrievals from the MOPITT satellite instrument
(panel a), averaged for each April in the period 2003 to 2012 (Deeter et al., 2014). The model results as well as the model
biases are shown for April 2016. The CO model estimates using C-CC (panel d) are characterized by a bias of -9×10^{17}
molec/cm^2 in the Northern Hemisphere (panel e), consistent with previous evaluations of CAM-chem (Emmons et al., 2020).
In C-GC (panel b), a negative bias still exists in the Northern Hemisphere (panel c), but is smaller at -5×10^{17} molec/cm^2. Across
all three model configurations a north-south gradient is observed in the model bias, with the bias in the Southern Hemisphere
being approximately 10^{18} molec/cm^2, which is of greater magnitude than the (negative) bias in the Northern Hemisphere. The
results from S-GC (panel f) are nearly identical to those in C-GC, with a smaller negative bias in the Northern Hemisphere
(panel g) than C-CC, but a larger positive bias in the Southern Hemisphere.

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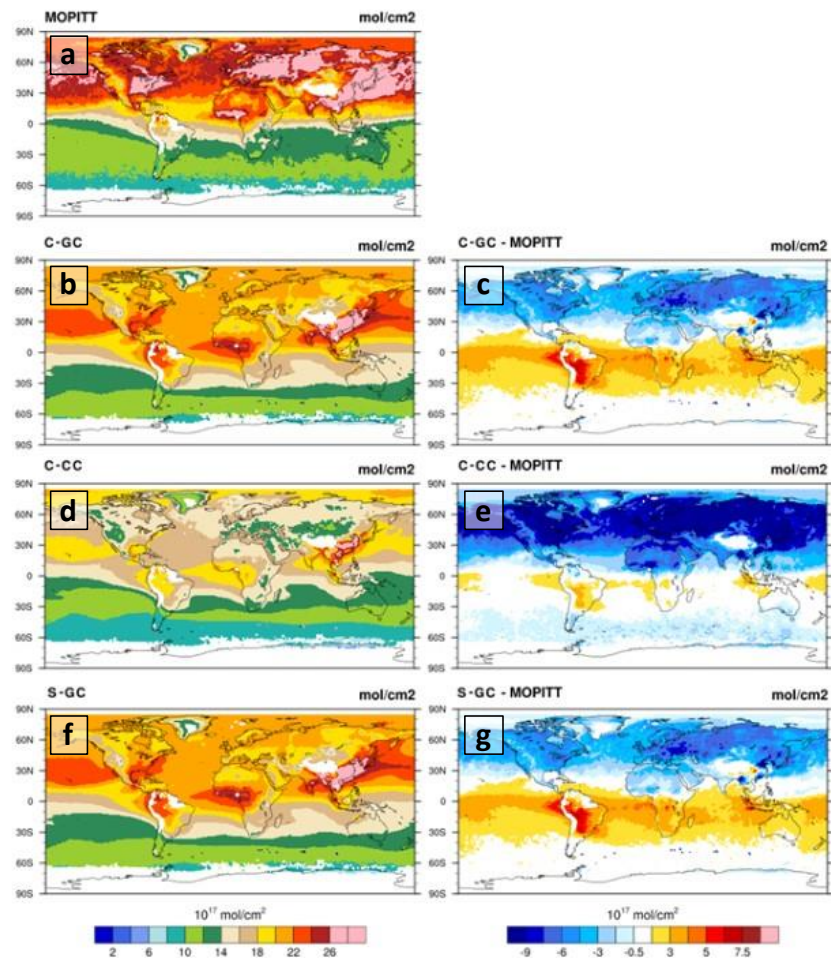
Figure 21. Total CO column in molec/cm^2 as observed by MOPITT during April for the 2003-2012 time period. (panel a), compared to the results from C-GC (2nd row, panels b and c), C-CC (3rd row, panels d and e), and S-GC (4th row, panels f and g) for April 2016. The measurements and model results are presented on the left (a, b, d, f), while the model biases are shown on the right (c, e, g).

5.4.   Wet and dry deposition tendencies

Finally, we compare simulated and observed surface deposition. Since deposition is the primary removal mechanism for atmospheric reactive nitrogen and sulfur species, the ability of a model to reproduce observed patterns of deposition provides an aggregate diagnostic for its representation of emissions, atmospheric chemistry, and the physical deposition processes.

Recent measurements have provided wet deposition rates in numerous geographical locations for the years 2005 to 2007 (Vet et al., 2014). Dry deposition fluxes are available from the same study but are limited to sulfur and nitrogen species. They are also limited to fewer geographical locations. Nonetheless we compare results from all three model configurations to the results from Vet et al. (2014) below.

986   Figure 22 compares the model-evaluated wet deposition rates of nitrogen at the surface for C-GC, C-CC, and S-GC. The total

987   nitrogen flux is calculated by adding surface fluxes from each individual nitrogen compound undergoing wet deposition.

988   Rainwater composition measurements are also displayed where available for comparison. We find correlation coefficients of

989   0.65, 0.66, and 0.67 for C-GC, C-CC and S-GC respectively with these observations. On average, the results from C-GC (panel

990   b) are closest to parity with a slope of 0.6, compared to 0.5 and 0.49 for C-CC (panel d) and S-GC (panel f). We do not find

991   any clear trends by location between the three models.

992

994   *Figure 22. Geographical distribution of the wet deposition flux of nitrogen for C-GC (top, panels a-b), S-GC (middle, panels c-d), and C-*
995   *CC (bottom, panels e-f). The annual mean value is shown as a map for each model, with circles used to indicate observational*
996   *measurements (left, panels a, c, e). A parity plot of the results against the rainwater composition measurements is also provided for each*
997   *model simulation (right, panels b, d, f).*

998

999 Comparing the dry deposition flux of nitrogen species at the surface from C-GC, C-CC and S-GC to in-situ measurements
1000 over North America (the only region present in this data set) from 2005 to 2007 shows that all models have positive biases.
1001 Relative to an observational mean of 1.57 kgN/ha/yr, C-GC has the best performance with a mean bias of +0.94 kgN/ha/yr,
1002 compared to +1.76 and +2.32 kgN/ha/yr from S-GC and C-CC respectively. These biases from all three models can be
1003 explained by either larger concentrations of nitrogen compounds or enhanced dry deposition velocities. However, we do not
1004 compensate for changes in nitrogen emissions between the time of the observations (2005-2007) and the simulated period,
1005 during which NO_x emissions are estimated to have increased (Emmons et al., 2020).

1006

1007 Figure 23 displays the evaluated wet deposition rates of non-sea salt sulfur from C-GC, C-CC, and S-GC alongside
1008 measurements of sulfur in rainwater for 2005. When comparing across model results, we find a global mean deposition rate of
1009 0.58, 0.38, and 0.50 kg S/ha/year in C-GC, S-GC, and C-CC respectively (computed as the global surface average of the left
1010 panels in Figure 23). The results from C-GC and C-CC show a correlation coefficient greater than 0.95, whereas C-GC and S-
1011 GC results show a correlation coefficient of 0.88.

1012

1013 Comparing to observational data, we find a mean bias of –2.40 kg S/ha/year (C-CC and C-GC) and –2.76 kg S/ha/year (S-GC)
1014 between the simulation results and rainwater composition measurements. This bias is location-dependent, with simulated data
1015 for Asia showing a lower bias than North America or Europe. For instance, over North America, measurements indicate a
1016 mean sulfur wet deposition flux of approximately 5 kg S/ha/year (for the year 2005), while the results at the same stations are
1017 lower with the slope of the linear fit equal to 0.2, 0.1, and 0.2 for C-GC, S-GC and C-CC respectively. This can be explained
1018 by the reduction in the sulfur wet deposition surface flux over the past decades. Previous literature has found that the deposition
1019 rate of sulfur over the Eastern U.S. has been decreasing at a rate of 1 kg S/ha/year[2] since 1990, with 60% of the reduction being
1020 in wet deposition rates and 40% in dry deposition rates (Zhang et al., 2018). Similar findings have been suggested for wet
1021 deposition rates over Europe (Theobald et al., 2019). A similar, but more recent, decrease over Asia has also been observed
1022 (Aas et al., 2019).

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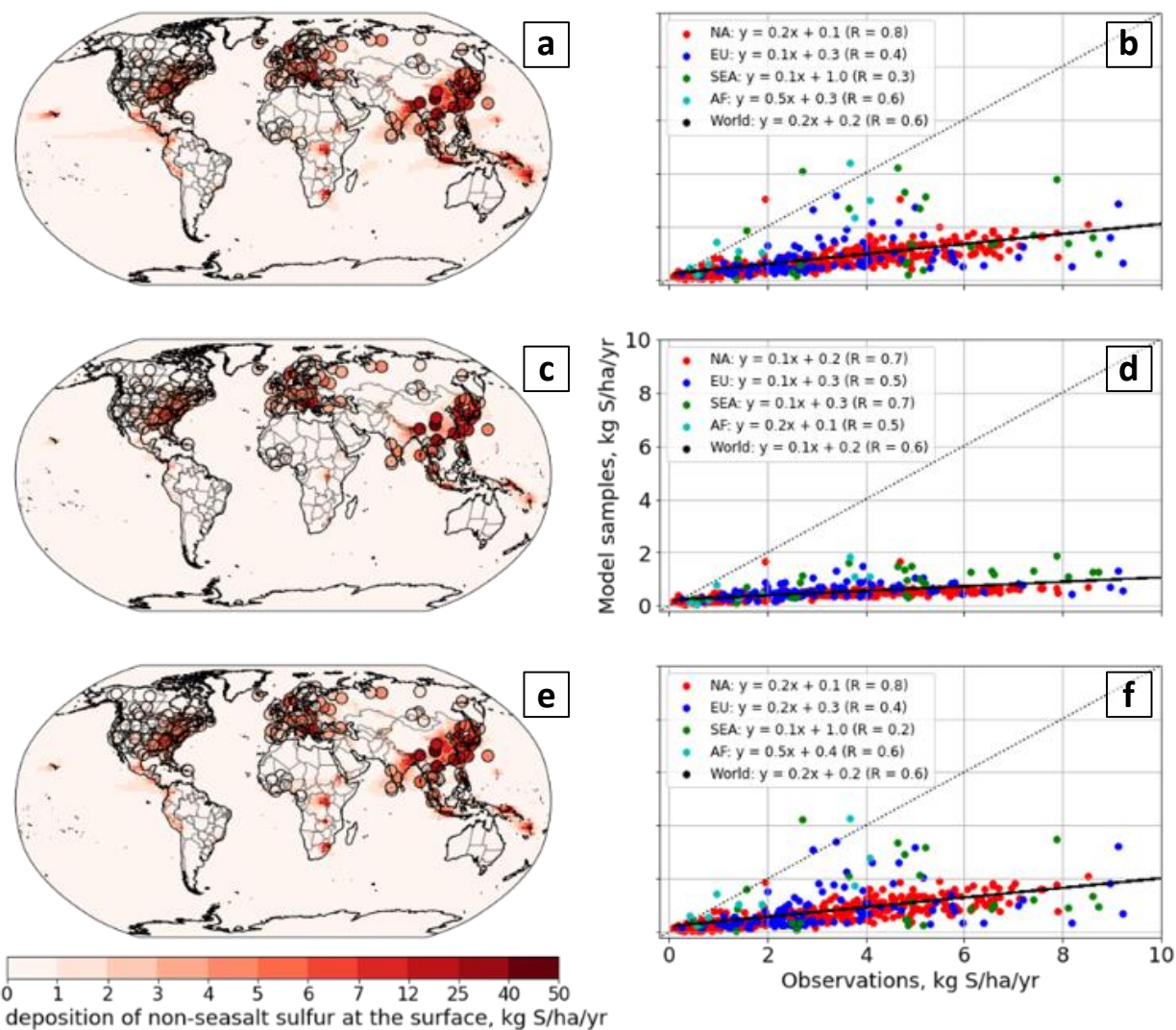
1024

1025 *Figure 23. As in Figure 22, but now for non-sea salt sulfur. Rows: C-GC (top), S-GC (middle), and C-CC (bottom).*

1026 It is difficult to say with confidence that the calculated bias is purely due to lack of recent data without new measurements to

1027 support this conclusion. However, our results do show that the simulation of sulfur deposition in C-GC more closely follows

1028 that in C-CC than that in S-GC. This could be due to either the simulated distribution of precipitation, the representation of

1029 aerosol, or the representation of scavenging processes, all of which differ between C-GC (or C-CC) and S-GC.

6.   Discussion and conclusion

We present the first implementation of the GEOS-Chem chemistry mechanism as an option in the Community Earth System Model (CESM). In addition to allowing users of CESM to take advantage of advancements in atmospheric modeling implemented in the GEOS-Chem model, this also allows the community to better understand why models disagree and how progress might be made to improve model performance and accuracy.

Our results suggest that differences in the representation of tropospheric halogen chemistry – in particular the representation and magnitude of emissions of short-lived bromine and chlorine sources – may be responsible for differences in simulated ozone between these model configurations. However, in addition to the recognized differences in chemical mechanisms, subtle structural differences in atmospheric models may have a significant role. Our evaluation of tropospheric ozone concentrations suggests that one of the key drivers in differences between CAM-chem and GEOS-Chem ozone fields differences may be differences in free tropospheric water vapor. Similarly, we show that sulfur deposition rates are approximately twice as great when running GEOS-Chem in a standalone model as opposed to running GEOS-Chem embedded in CESM, despite the use of identical emissions.

We also find that differences in the representation of wet scavenging are a significant contributor to differences in reactive nitrogen and halogen species distributions between GEOS-Chem and CAM-chem. The unification of convective transport and scavenging in GEOS-Chem helps to prevent movement of soluble species to the upper troposphere through convective updrafts, and therefore limits the effect of near surface halogen emissions from sea salt on ozone at higher altitudes.

Our implementation of GEOS-Chem in CESM is now publicly available for use. We envision that this model can become a powerful tool for research, forecast, and regulatory applications of global atmospheric chemistry, air quality, and climate research. Furthermore, this is also an important step towards the Multiscale Infrastructure for Chemistry and Aerosols (MUSICA), and thereof a truly modular Earth system model. By enabling us to fairly compare models down to individual processes, we can begin to understand precisely why different models perform better or worse in reproducing different measurements and accelerate our efforts to improve atmospheric modeling fidelity as a whole.

Finally, this work will foster collaboration between the GEOS-Chem and CESM-CAM-chem communities. GEOS-Chem is presently used and developed worldwide for research by over 100 university groups and government agencies, and CAM-chem similarly has numerous users and developers. The availability of GEOS-Chem as an option in CESM will stimulate broader interest in the GEOS-Chem community to use CESM, and in the CESM community to use GEOS-Chem. Indeed, we expect that on-line simulation of atmospheric chemistry will become increasingly attractive to GEOS-Chem users as the resolution of dynamical models increase, and that CESM will provide the principal vehicle for this because of its public

availability and support. By enabling improvements developed for GEOS-Chem to percolate into CESM without the need for re-implementation, this work will accelerate progress in atmospheric chemistry and Earth system modeling.

Author contributions

TMF, SDE, HL, and EWL were responsible for the software development. TMF performed the investigation, formal analysis, and validation. SDE, LKE, SRHB, and DJJ conceived of the project and acquired funding. SDE, LKE, SG, SRHB, and DJJ supervised the work. TMF performed all visualization and prepared the original draft. Review and editing were performed by all co-authors. All contributions are defined according to the CRediT taxonomy (https://casrai.org/credit).

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Code availability statement

GEOS-Chem as an option within CESM is currently being implemented into the CESM main branch, such that no additional download will be needed to use it. However, a standalone copy of the specific implementation of CESM including GEOS-Chem which was used to generate the results in this manuscript is permanently archived at https://github.com/CESM-GC/CESM-GC-Standalone/releases/tag/v1.0.0_review (permanent DOI: https://doi.org/10.5281/zenodo.6465076). To reproduce the results of this work, the repository should be used as-is without using features such as checkout_externals to acquire any additional code.

Data availability statement

Output data from the model are available from the authors upon request. Satellite data retrievals including from OMI/MLS and of carbon monoxide from the Terra/MOPITT instrument are available from the NASA Earth data archive (https://earthdata.nasa.gov; ftp://l5ftl01.larc.nasa.gov/MOPITT/). Surface observations of carbon monoxide are available from

Competing interests

The authors declare that they have no conflict of interest.

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